PHASE TRANSITIONS IN RANDOM MEDIA

by

Norman Benjamin Murphy

A dissertation submitted to the faculty of
The University of Utah
in partial fulfillment of the requirements for the degree of

Doctor of Philosophy

Department of Mathematics

The University of Utah

August 2012

The University of Utah Graduate School

STATEMENT OF DISSERTATION APPROVAL

The dissertation of **Norman Benjamin Murphy**

has been approved by the following supervisory committee members:

Kenneth M. Golden	, Chair	**June 15, 2012** Date Approved
Elena Cherkaev	, Member	**June 15, 2012** Date Approved
David C. Dobson	, Member	**June 15, 2012** Date Approved
Firas Rassoul-Agha	, Member	**June 15, 2012** Date Approved
Jingyi Zhu	, Member	**June 15, 2012** Date Approved

and by **Peter E. Trapa**, Chair of

the Department of **Mathematics**

and by Charles A. Wight, Dean of The Graduate School.

ABSTRACT

Transport in disordered composite media is a problem that arises throughout the sciences and engineering and has attracted significant theoretical, computational, and experimental interest. One of the key features of these types of problems is the critical dependence of the effective transport properties on system parameters, such as volume fraction, component contrast ratio, applied field strength, etc. In recent years a broad range of mathematical techniques have been developed to study phase transitions exhibited by such composites, revealing features which are virtually ubiquitous in disordered systems. Here we construct a multifaceted mathematical framework describing phase transitions exhibited by two phase random media, using techniques from: statistical mechanics, percolation theory, random matrix theory, and a critical theory for Stieltjes functions of a complex variable involving the spectral measure of a self-adjoint random operator (or matrix). In particular, we present a general theory for critical behavior of transport in two phase random media. The theory holds for lattice and continuum percolation models in both the static case with real parameters and the frequency dependent quasi-static case with complex parameters. Through a direct, analytic correspondence between the magnetization of the Ising model and the effective parameter problem of two phase random media, we show that the critical exponents of the transport coefficients satisfy the standard scaling relations for phase transitions in statistical mechanics. Our work also shows that delta components form in the underlying spectral measures at the spectral endpoints precisely at the percolation threshold p_c and $1 - p_c$. This is analogous to the Lee–Yang–Ruelle characterization of the Ising model phase transition, and identifies these transport transitions with the collapse of spectral gaps in these measures. Using random matrix theory, we also characterize these transport transitions via transitions in the eigenvalue statistics of the underlying random matrix. Finally, we construct a canonical ensemble statistical mechanics framework for general transport models of two phase random dielectric media, which parallels the Ising model. Our physically consistent model is formulated from first principles in physics, and is both physically transparent and mathematically tractable.

To my sweet sister Rachael whose last words were to me: “finish”

CONTENTS

LIST OF FIGURES

CHAPTER 1

INTRODUCTION

Composite media arise naturally throughout the physical and biological sciences, and are employed in a broad range of engineering and technological applications. The behavior of those media exhibiting a critical transition as system parameters are varied is particularly challenging to describe physically, and to predict mathematically. We now introduce examples of such media that have motivated much of the work presented in this dissertation, and review work that does not appear here.

Electrorheological (ER) fluids, which are suspensions of spherical particles in a dielectric liquid host, exhibit a liquid to solid phase transition as the strength E_0 of an applied electric field surpasses a critical value E_c [70, 120, 127, 128]. As the field increases, plastic or glass dielectric spheres, with diameters on the scale of tens of microns, form clusters and then chains which coalesce into columns with periodic lattice arrangements of the spheres, as shown in Figure 1.1 (a). Metal spheres form fractal net structures as shown in Figure 1.1 (b). This system undergoes an electrically-induced transition in the connectedness of the spheres. With an increase in viscosity of these suspensions by several orders of magnitude within a few milliseconds [126], they have been used in clutches, brakes, micro-fluidic valves, human prosthetics, and active mechanical elements capable of responding to environmental variations [120].

Another composite which displays complex critical behavior is sea ice, consisting of pure ice with submillimeter brine inclusions, as shown in Figure 1.1 (c), whose volume fraction ϕ, geometry, and connectedness vary significantly with temperature T. The polar ice packs are both indicators and agents of climate change. They also host extensive algal and bacterial communities, which live in the brine inclusions and sustain life in the polar oceans. Fluid flow through porous sea ice mediates a broad range of processes such as the growth and decay of seasonal ice, the evolution of surface melt ponds and ice pack reflectance, and biomass build-up [61]. In [62] an *on-off switch* was identified for fluid transport in sea ice. For brine volume fractions ϕ below about 5%, columnar sea ice is effectively impermeable to fluid flow, while for ϕ above 5%, it is increasingly permeable. This critical brine volume fraction $\phi_c \approx 5\%$ corresponds to a critical temperature $T_c \approx -5^\circ$ C for a typical bulk sea ice salinity of 5 parts per thousand, which is known as the *rule of fives*. Fluid flow is facilitated by *brine channels* − connected brine structures ranging in scale from a few centimeters for horizontal cross-sections to a meter or more in the vertical direction, as shown in Figure 1.1 (d). The critical behavior of fluid flow through sea ice was postulated in [62] to result from a temperature-driven transition in connectedness of the brine microstructure, or a *percolation threshold*. The 5% critical brine volume fraction in sea ice was predicted by noting the close similarity of its microstructure to that of stealthy, radar absorbing materials, and adapted for sea ice a continuum percolation model for compressed powders [78, 79] used in the design of these materials.

Bone also displays a complex, porous microstructure whose characteristics depend on

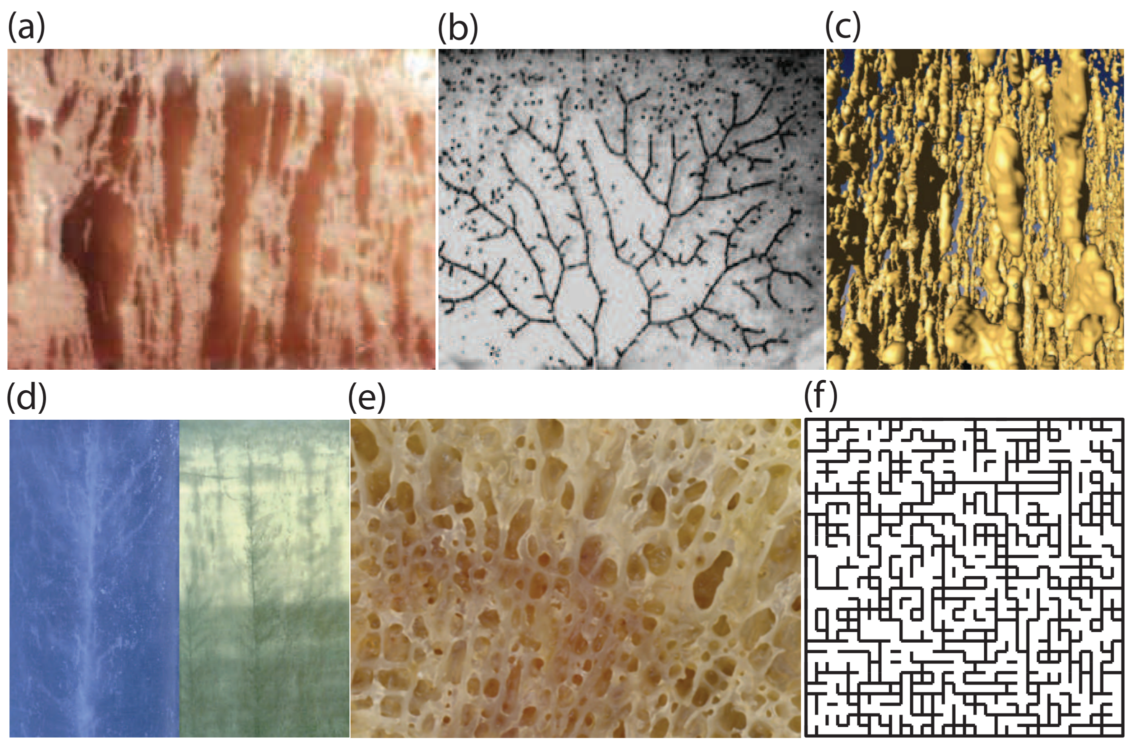

Figure 1.1. Systems which undergo phase transitions. (a) Columnar ground states of dielectric spheres in an ER fluid (Martin Whittle). (b) Fractal network of metal spheres in an ER fluid (Weijia Wen). (c) X-ray CT volume rendering of the brine phase within a lab-grown sea-ice single crystal (Golden et al. [63]). (d) Vertical brine channels (Kenneth. M. Golden (left) and David M. Cole (right)). (e) The porous microstructure in a proximal femur (Maria–Grazia Ascenzi). (f) A realization of the two-dimensional lattice percolation model.

its macrostructure. The strength of bone and its ability to resist fracture depend strongly on a porous microstructure, like that shown in Figure 1.1 (e), and in particular, on the *quality* of the connectedness of the hard, solid phase. Osteoporotic cancellous bone can become more disconnected and remaining connections can become more tenuous or fragile. It is crucial that *nondestructive* methods of analyzing bone microstructure are developed in order to facilitate the comfort and long term well being of patients stricken with this crippling disease. In [64] we applied spectral methods to investigate osteoporosis-driven transitions in trabecular bone microstructure. This introduces important nondestructive methods of analyzing bone microstructure and its properties, which may eventually help in clinical applications.

Lattice and continuum percolation models have been used to study a broad range of materials including rocks [20, 21], semiconductors [113], thin films [38], glacial ice [53], bone [64, 109], polycrystalline metals [30], radar absorbing coatings [79], carbon nanotube composites [80], and sea ice [62, 63]. In the simplest case of the two-dimensional square lattice [116, 122], as shown in Figure 1.1 (f), the bonds are open with probability p and closed with probability $1-p$. Connected sets of open bonds are called open clusters. The average cluster size grows as p increases, and there is a critical probability p_c, $0 < p_c < 1$, called the *percolation threshold*, where an infinite cluster of open bonds first appears. In dimension $d = 2$, $p_c = 1/2$, and in $d = 3$, $p_c \approx 0.25$. Now consider transport through the associated *random resistor network* (RRN), where the bonds are assigned electrical conductivities σ_1 with probability $1-p$, and σ_2 with probability p. The effective conductivity σ^* exhibits critical behavior as $\sigma_1 \longrightarrow 0$, $\sigma^* = 0$ for $p < p_c$ while $\sigma^* > 0$ for $p > p_c$, with $\sigma^*(p, 0) \sim (p - p_c)^t$, as $p \longrightarrow p_c^+$, where t is the conductivity critical exponent. believed to be *universal* for lattices depending only on dimension.

The critical behavior of the random resistor network is reminiscent of a phase transition in statistical mechanics [13, 37, 52, 72], like that exhibited by an Ising ferromagnet [36, 121] around its Curie point at a critical temperature $T = T_c$, as the applied magnetic field strength $H \longrightarrow 0$. The formulations and basic physics of these two classes of problems are nevertheless quite different. In [59, 60], however, it was observed that the *analytic continuation method* (ACM) for bounding effective transport properties of composites [11, 65, 86] provides a mathematical link between them, through the Lee–Yang Theorem in statistical mechanics [3, 4, 81]. This theorem, which states that the zeros of the partition function lie on the unit circle in the activity variable, yields a logarithmic integral for the free energy of an Ising model, and a Stieltjes integral representation for the magnetization $M(H)$. In the ACM, $m = \sigma^*/\sigma_2$ is an analytic function of h, taking the upper half plane to the upper half plane. A Stieltjes integral representation for $F(s) = 1 - m(h)$, where $s = 1/(1-h)$, is used to obtain rigorous bounds on σ^* given microstructural information. This formula is based on a resolvent representation for the electric field, involving a self-adjoint random operator (or matrix) M_1, say, not to be confused with magnetization. All the geometry of the composite is incorporated through a spectral measure μ of this operator. Through these *mathematical* parallels the two classes of problems can be placed on an equal footing.

This analytic parallel has been exploited to show that the critical exponents of transport in both lattice and continuum percolation models obey the classical scaling relations of statistical mechanics [60, 89]. Moreover, as in the Ising critical transition [106], we have shown [89] that the critical behavior in these models is due to the collapse of *spectral gaps* of the underlying spectral measure, and the subsequent formation of delta components at the spectral endpoints. This theory holds for lattice and continuum percolation models in both the static case with real parameters and the frequency dependent quasi-static case with complex parameters [89]. It has also been extended from insulator/conductor systems to conductor/superconductor systems [89]. These results help lay the groundwork for the analysis of sea ice permittivity data collected in the polar regions. Moreover, it can potentially be used to monitor changes in the microstructure, the fluid transport properties, and the biogeophysical processes that are controlled by fluid flow, by *remotely* monitoring the effective electromagnetic properties of sea ice, such as its effective complex permittivity ϵ^*. The general theory of spectral representations for effective parameters and system energy, and the spectral characterization of critical behavior in transport is the topic of Chapter 2. While critical behavior of percolation models of two-phase random media is discussed in Chapter 3.

On *finite* two-component RRN, the operator M_1 is a real symmetric random matrix. In this case, detailed information regarding transport transitions may be gleaned from the eigenvalue statistics of this matrix [88]. In Chapter 5 we study transport transitions of various finite RRN percolation models using techniques of random matrix theory (RMT). As a function of the volume fraction p, we demonstrate that the eigenvalue spacing statistics, and the well known RMT eigenvalue statistics Σ^2 and Δ_3, of M_1 have a transitional behavior much like that of the Anderson transition of mesoscopic and quantum conductors [117]. Our results illustrate strikingly, that the spectrum associated with macroscopic electrical systems exhibit the same universal fluctuations as the energy spectrum of mesoscopic electrical systems of size 10^{-6}m and quantum systems nine orders of magnitude smaller.

In [90, 92] we calculated the spectral measures and random matrix statistics for brine channels, melt pond networks, and large scale ice floes. To our knowledge, this is the first time that spectral measures and random matrix statistics of sea ice structures have been calculated. Our results provide a new way of obtaining important structural information that can eventually give key parameters in climate models through novel techniques of *upscaling*, aiding the prediction and understanding of climate change.

The theory of polynomials orthogonal to a measure naturally arises in the ACM through Padé approximants $F_n = P_n^{[1]}/P_n$ of $F(s;\mu)$, a ratio of polynomials with exponential convergence as $n \longrightarrow \infty$ [125]. Both the denominator P_n and the numerator $P_n^{[1]}$ are examples of "generalized numerator polynomials" $\{P_n^{[i]}\}_{n=0}^{\infty}$ of order $i = 0, 1, \ldots$, which are orthogonal with respect to a measure $\mu^{[i]}$, where $P_n^{[0]} = P_n$ and $\mu^{[0]} = \mu$ [57, 125]. They satisfy a three-term recursion relation [57, 124] which may be written as $J^{[i]}\vec{P}^{[i]} = \lambda\vec{P}^{[i]}$, where $\vec{P}^{[i]} = (P_0^{[i]}, P_1^{[i]}, \ldots)$ and $J^{[i]}$ is an *infinite* tridiagonal Jacobi matrix [118]. In finite RRN, the operators M_1 and $J^{[i]}$ are *finite* real symmetric random matrices of size N, say, and eigenvalues of $J^{[i]}$ are the roots of the polynomial $P_{N-i}^{[i]}$. Moreover, the roots of $P_N^{[0]} = P_N$ are the eigenvalues of $J^{[0]}$ *and* M_1 [57]. This theory, presented in Chapter 4, gives physical significance for the Stieltjes transforms $F^{[i]}(s;\mu^{[i]})$ of the measures $\mu^{[i]}$. We also provide a closed form solution for the moments $\mu_j^{[i]}$ of $\mu^{[i]}$ in terms of the moments μ_j of μ.

In [91] we constructed a canonical ensemble statistical mechanics framework for general transport models of two-phase random dielectric media, and used it to study general features of ER fluids. In this model, we incorporate a detailed decomposition of the system energy, in terms of Herglotz functions involving μ, which *separates* parameter information in s and E_0 from complicated geometric interactions incorporated in the measure μ. These energy representations are *exact* in the infinite volume limit. Due to this parameter separation property, the physically consistent model is mathematically tractable, physically transparent, and closely parallels the Ising model. This mathematical framework is the topic of Chapter 6.

In order to motivate the effective parameter problem for general two-phase stationary conductive media in lattice and continuum settings, given in Section 2.1, we first analyze canonical examples of two-component RRN in Section 1.1, and two-phase continuum composite microstructures in Section 1.2. There, we derive integral representations for the associated effective parameters. The analysis of these simple lattice and continuum composites reveals important features of the effective parameters of general two-component stationary random media.

1.1 Hierarchical Series-Parallel Random Resistor Network

In this section we derive an integral representation for the effective conductance σ^* of hierarchical series-parallel RRN, involving a positive, discrete measure. This representation for σ^* was originally introduced by Bergman and Milton [11, 86], and was later shown by Golden and Papanicolaou [65] to have theoretical foundations in the functional analysis of bounded linear operators. In this dissertation, we demonstrate that this representation has deep, far reaching physical and mathematical implications.

Consider a RRN of n resistors of resistance $R_j = 1/\sigma_j$, $j = 1, \ldots, n$, where σ_j is the conductance of resistor j, which randomly takes values σ_1 and σ_2 in the proportions $p_1 = (n-k)/n$ and $p_2 = k/n$, respectively. Here, $k = 0, \ldots, n$, $p_1 + p_2 = 1$, and σ_j for $j = 1, 2$ is not to be confused with the *values* σ_1 and σ_2. Let Ω be the space of all $2^n n!$ permutations of the set $\{\sigma_j\}_{j=1}^n$ of binary conductances, and let $\Omega_p \subset \Omega$ be those which satisfy the constraint $p_1 + p_2 = 1$.

We focus on RRN which are wired in a hierarchical series-parallel topology. More specifically, consider the networks shown in Figure 1.2. We will call these 0^{th} order hierarchical series-parallel network. We define 1^{st} order hierarchical series-parallel network as follows. First, consider the network of parallel conductors shown in Figure 1.2 (a) with conductor σ_l replaced by n_l conductors wired in series, where $l = 1, \ldots, L$. For every $l = 1, \ldots, L$ and $j = n_{l-1} + 1, \ldots, n_l$, with $n_0 = 0$, the conductances σ_j randomly take values σ_1 and σ_2 in the proportions $p_{1,l} = (n_l - k_l)/n_l$ and $p_{2,l} = k_l/n_l$, respectively, with $\sum_{l=1}^L k_l = k$ and $\sum_{l=1}^L n_l = n$. For simplicity, we have suppressed the notation $L = L(\omega)$, $k_l = k_l(\omega)$ and $n_l = n_l(\omega)$, where $\omega \in \Omega_p$. Second, consider the network of series conductors shown in Figure 1.2 (b) with conductor σ_l replaced by n_l conductors wired in parallel, $l = 1, \ldots, L$, with the σ_j randomly determined as before. In this way, we iteratively construct M^{th} order hierarchical series-parallel RRN. Theorem 1 displays the main result of this section.

Theorem 1 *Consider a M^{th} order hierarchical series-parallel RRN, as described above, subject to a (constant) voltage potential V_0. For each $\omega \in \Omega_p$, there exists a positive measure*

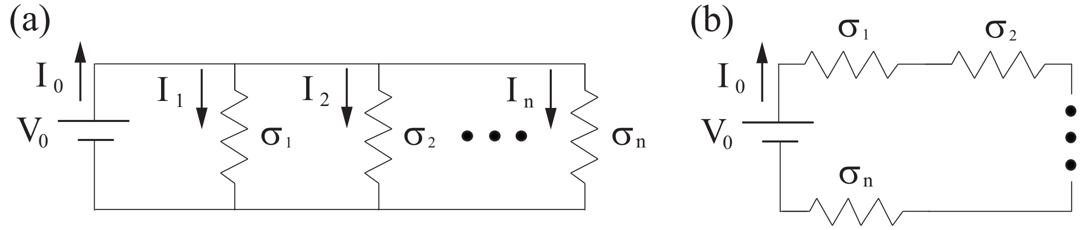

Figure 1.2. (a) RRN of parallel resistors subject to a voltage V_0 and total current I_0. (b) RRN of series resistors subject to a voltage V_0 and total current I_0.

μ_ω supported on $[0,1]$ and a number N_ω such that the effective conductance σ^*_ω and the power $\mathcal{H}_\omega$ dissipated by the network satisfies

$$\mathcal{H}_\omega = \frac{V_0^2\sigma^*_\omega}{N_\omega}, \quad \sigma^*_\omega = \sigma_2(1-F(s;\mu_\omega)), \quad F(s;\mu_\omega) = \int_0^1 \frac{d\mu_\omega(\lambda)}{s-\lambda}, \tag{1.1}$$

$$\mu_\omega(d\lambda) = \sum_{l=1}^{L_0} m_l\delta_{\lambda_l}(d\lambda), \quad 0 \le \lambda_l(\omega) \le 1, \quad m_l(\omega) \ge 0$$

where $s = 1/(1-\sigma_1/\sigma_2)$, $V_0, N_\omega > 0$, $L_0 = L_0(\omega)$, and $\delta_{\lambda_l}(d\lambda)$ is the delta measure concentrated at λ_l.

Proof: We will prove Theorem 1 inductively. First, let $\omega \in \Omega_p$ be a statistical realization of the 0^{th} order RRN shown in Figure 1.2 (a), subject to a voltage potential V_0. Here σ_j is the conductance of resistor j, which randomly take values σ_1 and σ_2 in the proportions $p_1 = (n-k)/n$ and $p_2 = k/n$, respectively, where $k = 0,\dots,n$ and $p_1+p_2 = 1$. By Kirchoff's voltage law, the voltage drop V_j across each resistor is V_0 so that $V_j = V_0$ for all $j = 1,\dots,n$. The electrical current I_j through resistor j is given by Ohm's law, $I_j = \sigma_j V_j$, and the power dissipated by resistor j is given by $I_jV_j = \sigma_j V_j^2$. The total power $\mathcal{H}_\omega$ dissipated by the RRN is thus

$$\mathcal{H}_\omega = \sum_{j=1}^n \sigma_j V_j^2 = V_0^2\sigma_{\text{eff}}, \quad \sigma_{\text{eff}} = \sum_{j=1}^n \sigma_j, \tag{1.2}$$

where σ_{eff}/n is the *arithmetic mean* of the $\{\sigma_j\}$. In the variable $s = 1/(1-\sigma_1/\sigma_2)$,

$$\frac{\sigma_{\text{eff}}}{\sigma_2} = \frac{1}{\sigma_2}\sum_{j=1}^n \sigma_j = \frac{1}{\sigma_2}((n-k)\sigma_1 + k\sigma_2) = n\left(p_1\left(1-\frac{1}{s}\right)+p_2\right) = n\left(1-\frac{p_1}{s}\right). \tag{1.3}$$

Therefore in (1.1), $N_\omega = 1/n$, $\sigma^*_\omega = \sigma_2(1-p_1/s)$, $L_0 = 1$, and $\mu_\omega(d\lambda) = m_1\delta_{\lambda_1}(d\lambda)$, where $m_1 = p_1$ and $\lambda_1 = 0$ satisfy $0 \le m_1, \lambda_1 \le 1$. We note that, for this simple resistor topology, σ^*_ω is invariant under permutations of the $\{\sigma_j\}$, and the randomness plays no role.

Second, let $\omega \in \Omega_p$ be a statistical realization of the 0^{th} order RRN shown in Figure 1.2 (b), subject to a voltage potential V_0, with the σ_j randomly determined as before. By Kirchoff's current law and Ohm's law, the electrical current through each resistor is $I_j = \sigma_j V_j = I_0$ for all $j = 1, \dots, n$, where I_0 is the (constant) total electrical current generated by the battery supplying the voltage potential V_0 to the RRN. Kirchoff's voltage law now yields

$$V_0 = \sum_{j=1}^{n} V_j = I_0 \sum_{j=1}^{n} \sigma_j^{-1} = \frac{I_0}{\sigma_{\text{eff}}}, \quad \sigma_{\text{eff}} = \left(\sum_{i=1}^{n} \sigma_i^{-1} \right)^{-1}, \tag{1.4}$$

where σ_{eff}/n is the *harmonic mean* of the $\{\sigma_j\}$. Together, we have $V_j = I_0/\sigma_j = V_0\sigma_{\text{eff}}/\sigma_j$. This implies that the total power $\mathcal{H}_\omega$ dissipated by the RRN is given by

$$\mathcal{H}_\omega = \sum_{j=1}^{n} \sigma_j V_j^2 = (V_0\sigma_{\text{eff}})^2 \sum_{i=1}^{n} \sigma_i^{-1} = V_0^2 \sigma_{\text{eff}}, \tag{1.5}$$

In the variable s we have

$$\begin{aligned} \frac{\sigma_{\text{eff}}}{\sigma_2} = \frac{1}{\sigma_2} \left(\sum_{j=1}^{n} \sigma_j^{-1} \right)^{-1} &= \frac{1}{\sigma_2} \left(\frac{n-k}{\sigma_1} + \frac{k}{\sigma_2} \right)^{-1} = \frac{1}{n} \left(p_1 \left(1 - \frac{1}{1-s} \right) + p_2 \right)^{-1} \\ = \frac{1}{n} \left(1 - \frac{p_1}{1-s} \right)^{-1} &= \frac{1}{n} \frac{s-1}{s-p_2} = \frac{1}{n} \left(1 - \frac{p_1}{s-p_2} \right) \end{aligned} \tag{1.6}$$

Therefore in (1.1), $N_\omega = n$, $\sigma_\omega^* = \sigma_2(1 - p_1/(s - p_2))$, $L_0 = 1$, and $\mu_\omega(d\lambda) = m_1\delta_{\lambda_1}(d\lambda)$, where $m_1 = p_1$ and $\lambda_1 = p_2$ satisfy $0 \leq m_1, \lambda_1 \leq 1$. As before, for this simple resistor topology, σ_ω^* is invariant under permutations of the $\{\sigma_j\}$, and the randomness plays no role.

We now verify equation (1.1) for 1^{st} order hierarchical series-parallel RRN. First, let $\omega \in \Omega_p$ be a statistical realization of the RRN shown in Figure 1.2 (a) with conductor σ_l replaced by n_l conductors wired in series, $l = 1, \dots, L$. For each $l = 1, \dots, L$ and $j = n_{l-1} + 1, \dots, n_l$, with $n_0 = 0$, the σ_j randomly take values σ_1 and σ_2 in the proportions $p_{1,l} = (n_l - k_l)/n_l$ and $p_{2,l} = k_l/n_l$, respectively, where $\sum_{l=1}^{L} k_l = k$, $\sum_{l=1}^{L} n_l = n$, and $p_{1,l} + p_{2,l} = 1$. For simplicity we suppress the notation $L = L(\omega)$, $k_l = k_l(\omega)$ and $n_l = n_l(\omega)$. In contrast to the network topologies considered above, the randomness of the $\{\sigma_j\}$ plays a key role here. By iterating our previous analysis of series and then parallel RRN, we find that the total power $\mathcal{H}_\omega$ dissipated by the RRN in configuration $\omega \in \Omega_p$ is given by

$$\mathcal{H}_\omega = V_0^2 \sigma_{\text{eff}}, \quad \sigma_{\text{eff}} = \sum_{l=1}^{L} \left(\sum_{j=n_{l-1}+1}^{n_l} \sigma_j^{-1} \right)^{-1}, \tag{1.7}$$

In the variable s, we have

$$\frac{\sigma_{\text{eff}}}{\sigma_2} = \frac{1}{\sigma_2}\sum_{l=1}^{L}\left(\frac{n_l-k_l}{\sigma_1}+\frac{k_l}{\sigma_2}\right)^{-1} = \sum_{l=1}^{L}\frac{1}{n_l}\left(p_{1,l}\left(1-\frac{1}{1-s}\right)+p_{2,l}\right)^{-1} \tag{1.8}$$
$$= \sum_{l=1}^{L}\frac{1}{n_l}\left(1-\frac{p_{1,l}}{1-s}\right)^{-1} = \sum_{l=1}^{L}\frac{1}{n_l}\,\frac{s-1}{s-p_{2,l}} = \sum_{l=1}^{L}\frac{1}{n_l}\,\left(1-\frac{p_{1,l}}{s-p_{2,l}}\right)$$
$$= \frac{1}{n_h^*}\left(1-\sum_{l=1}^{L}\frac{p_{1,l}\,n_h^*/n_l}{s-p_{2,l}}\right) = \frac{1}{n_h^*}\left(1-\sum_{l=1}^{L}\frac{m_l}{s-\lambda_l}\right),$$

where $n_h^* = (\sum_{l=1}^{L} n_l^{-1})^{-1}$ and n_h^*/L is the harmonic mean of the numbers $\{n_l\}$. Therefore in equation (1.1), $N_\omega = n_h^*$, $\sigma_\omega^* = \sigma_2(1-\sum_{l=1}^{L_0} m_l/(s-\lambda_l))$, $L_0 = L$, $\mu_\omega(d\lambda) = \sum_{l=1}^{L} m_l\delta_{\lambda_l}(d\lambda)$, $m_l = p_{1,l}\, n_h^*/n_l$, and $\lambda_l = p_{2,l}$. As $0 < n_h^*/n_l \le 1$ and $0 \le p_{2,l} \le 1$, we have $0 \le m_l, \lambda_l \le 1$.

Second, let $\omega \in \Omega_p$ be a statistical realization of the RRN shown in Figure 1.2 (b) with conductor σ_l replaced by n_l conductors wired in parallel, $l = 1, \ldots, L$. For every $l = 1, \ldots, L$ and $j = n_{l-1}+1, \ldots, n_l$, with $n_0 = 0$, the σ_j randomly take values σ_1 and σ_2 as before. By iterating our previous analysis of parallel then series RRN, we find that the total power $\mathcal{H}_\omega$ dissipated by the RRN in configuration $\omega \in \Omega_p$ is given by

$$\mathcal{H}_\omega = V_0^2\sigma_{\text{eff}}, \quad \sigma_{\text{eff}} = \left(\sum_{l=1}^{L}\left(\sum_{j=n_{l-1}+1}^{n_l}\sigma_j\right)^{-1}\right)^{-1}, \tag{1.9}$$

In the variable s, we have

$$\frac{\sigma_{\text{eff}}}{\sigma_2} = \frac{1}{\sigma_2}\left(\sum_{l=1}^{L}((n_l-k_l)\sigma_1+k_l\sigma_2)^{-1}\right)^{-1} = \left(\sum_{l=1}^{L}\frac{1}{n_l}\left(p_{1,l}\left(1-\frac{1}{s}\right)+p_{2,l}\right)^{-1}\right)^{-1}$$
$$= \left(\sum_{l=1}^{L}\frac{1}{n_l}\left(1-\frac{p_{1,l}}{s}\right)^{-1}\right)^{-1} = \left(\sum_{l=1}^{L}\frac{1}{n_l}\,\frac{s}{s-p_{1,l}}\right)^{-1} = \left(\sum_{l=1}^{L}\frac{1}{n_l}\,\left(1+\frac{p_{1,l}}{s-p_{1,l}}\right)\right)^{-1}$$
$$= n_h^*\left(1+\sum_{l=1}^{L}\frac{p_{1,l}\,n_h^*/n_l}{s-p_{1,l}}\right)^{-1} = n_h^*\left(1+\tilde{F}(s)\right)^{-1} = n_h^*\left(1-\frac{\tilde{F}(s)}{1+\tilde{F}(s)}\right)$$
$$= n_h^*(1-F(s)) = n_h^*\left(1-\sum_{l=1}^{L}\frac{m_l}{s-\lambda_l}\right). \tag{1.10}$$

Here we have defined

$$\tilde{F}(s) = \sum_{l=1}^{L}\frac{\tilde{m}_l}{s-\tilde{\lambda}_l} = \frac{Q_L(s)}{P_L(s)}, \qquad F(s) = \frac{\tilde{F}(s)}{1+\tilde{F}(s)} = \sum_{l=1}^{L}\frac{m_l}{s-\lambda_l} = \frac{Q_L(s)}{P_L(s)+Q_L(s)}$$
$$P_L(s) = \prod_{l=1}^{L}(s-\tilde{\lambda}_l), \qquad Q_L(s) = \sum_{l=1}^{L}\tilde{m}_l\prod_{l\neq j=1}^{L}(s-\tilde{\lambda}_j) = \sum_{l=1}^{L}m_l\prod_{l\neq j=1}^{L}(s-\lambda_l), \tag{1.11}$$

where $\tilde{\lambda}_l = p_{1,l}$ and $\tilde{m}_l = p_{1,l}\, n_h^*/n_l$. Therefore in equation (1.1), we have $N_\omega = 1/n_h^*$, $\sigma_\omega^* = \sigma_2(1 - \sum_{l=1}^{L_0} m_l/(s-\lambda_l))$, $L_0 = L$, $\mu_\omega(d\lambda) = \sum_{l=1}^{L} m_l \delta_{\lambda_l}(d\lambda)$, and the values of m_l and λ_l in (1.10) are determined as follows. The $\{\lambda_l\}$ are the roots of the polynomial $Q_L(s)+P_L(s)$ and, multiplying $F(s) = \sum_{l=1}^{L} m_l/(s-\lambda_l) = Q_L(s)/(Q_L(s)+P_L(s))$ by $s-\lambda_l$ and letting $s \longrightarrow \lambda_l$, L'Hôpital's rule shows that

$$m_l = \lim_{s \longrightarrow \lambda_l} (s-\lambda_l)F(s) = \frac{Q_L(\lambda_l)}{Q'(\lambda_l) + P_L'(\lambda_l)} \tag{1.12}$$

where the prime denotes differentiation in the variable s. We now prove that λ_l and m_l are real and satisfy $0 \le \lambda_l, m_l \le 1$.

The roots $\tilde{\lambda}_l = p_{1,l}$ of $P_L(s)$ and the numbers $\tilde{m}_l = p_{1,l}\, n_h^*/n_l$ are clearly real, and satisfy $0 \le \tilde{m}_l, \tilde{\lambda}_l \le 1$. However, as $\tilde{m}_l = 0$ when $\tilde{\lambda}_l = 0$ we may assume, without loss of generality, that we have $0 < \tilde{m}_l, \tilde{\lambda}_l \le 1$. Moreover by redefining the $\tilde{m}_l$ and $\tilde{m}_j$ for $l \ne j$ such that $p_{1,l} = p_{1,j}$, we may also assume, without loss of generality, that the $\{\tilde{\lambda}_l\}$ are distinct. Let's order them so that $\tilde{\lambda}_1 < \tilde{\lambda}_2 < \cdots < \tilde{\lambda}_L$. The roots of $Q_L(s)$ and $P_L(s)$ are the zeros and poles of $\tilde{F}(s)$, respectively. For $s < \tilde{\lambda}_1$ we have $\tilde{F}(s) < 0$, and for $s > \tilde{\lambda}_L$ we have $\tilde{F}(s) > 0$. As the $\tilde{m}_l$ are positive, the function $\tilde{F}(s)$ is monotonic decreasing: $\tilde{F}'(s) = -\sum_{l=1}^{L} \tilde{m}_l/(s-\tilde{\lambda}_l)^2 < 0$. This implies that $\tilde{F}(s)$ has precisely one root in each of the intervals $(\tilde{\lambda}_l, \tilde{\lambda}_{l+1})$, $l = 1, \ldots, L-1$. Therefore the roots and poles of $\tilde{F}(s)$ interlace, as shown in Figure 1.3. By equation (1.11), the roots of $\tilde{F}(s)$ and $F(s)$ coincide, and the poles λ_l of $F(s)$ are defined by $\tilde{F}(\lambda_l) = -1$. As $F'(s) = \tilde{F}'(s)/(1+\tilde{F}(s))^2 < 0$, the function $F(s)$ is monotonic decreasing. This implies that the poles λ_l of $F(s)$ satisfy $\lambda_l < \tilde{\lambda}_l < \lambda_{l+1} < \tilde{\lambda}_{l+1} \le 1$, $l = 1, \ldots, L-1$, with $\tilde{\lambda}_1 > 0$, which implies that the poles of $F(s)$ interlace the poles of $\tilde{F}(s)$ (see Figure 1.3). As $\tilde{F}(0) = -1$, we have $\lambda_1 = 0$:

$$\tilde{F}(0) = \sum_{l=1}^{L} \frac{p_{1,l}\, n_h^*/n_l}{-p_{1,l}} = -n_h^* \sum_{l=1}^{L} \frac{1}{n_l} = -1, \tag{1.13}$$

so that $0 \le \lambda_l \le 1$, $l = 1, \ldots, L$. The m_l satisfy $m_l = \lim_{s \longrightarrow \lambda_l}(s-\lambda_l)F(s)$, or

$$m_l = \lim_{s \longrightarrow \lambda_l} \frac{(s-\lambda_l)\tilde{F}(s)}{1+\tilde{F}(s)} = \lim_{s \longrightarrow \lambda_l} \frac{\tilde{F}(s) + (s-\lambda_l)\tilde{F}'(s)}{\tilde{F}'(s)} = \frac{\tilde{F}(\lambda_l)}{\tilde{F}'(\lambda_l)} = \frac{-1}{\tilde{F}'(\lambda_l)} \tag{1.14}$$

where we have used L'Hôpital's rule. As $0 \le \lambda_l < \tilde{\lambda}_l \le 1$ for all $l = 1, \ldots, L$, we have

$$-\tilde{F}'(\lambda_l) = \sum_{i=1}^{L} \frac{\tilde{m}_i}{(\lambda_l - \tilde{\lambda}_i)^2} > \sum_{i=1}^{L} \frac{\tilde{m}_i}{|\lambda_l - \tilde{\lambda}_i|} \ge \left| \sum_{i=1}^{L} \frac{\tilde{m}_i}{\lambda_l - \tilde{\lambda}_i} \right| = |\tilde{F}(\lambda_l)| = 1 \tag{1.15}$$

This implies that $m_l = -1/\tilde{F}'(\lambda_l) = \left(\sum_{i=1}^{L} \tilde{m}_i/(\lambda_l - \tilde{\lambda}_i)^2\right)^{-1}$ satisfies $0 \le m_l \le 1$. This concludes our verification of equation (1.1) for 1st order hierarchical series-parallel RRN. It

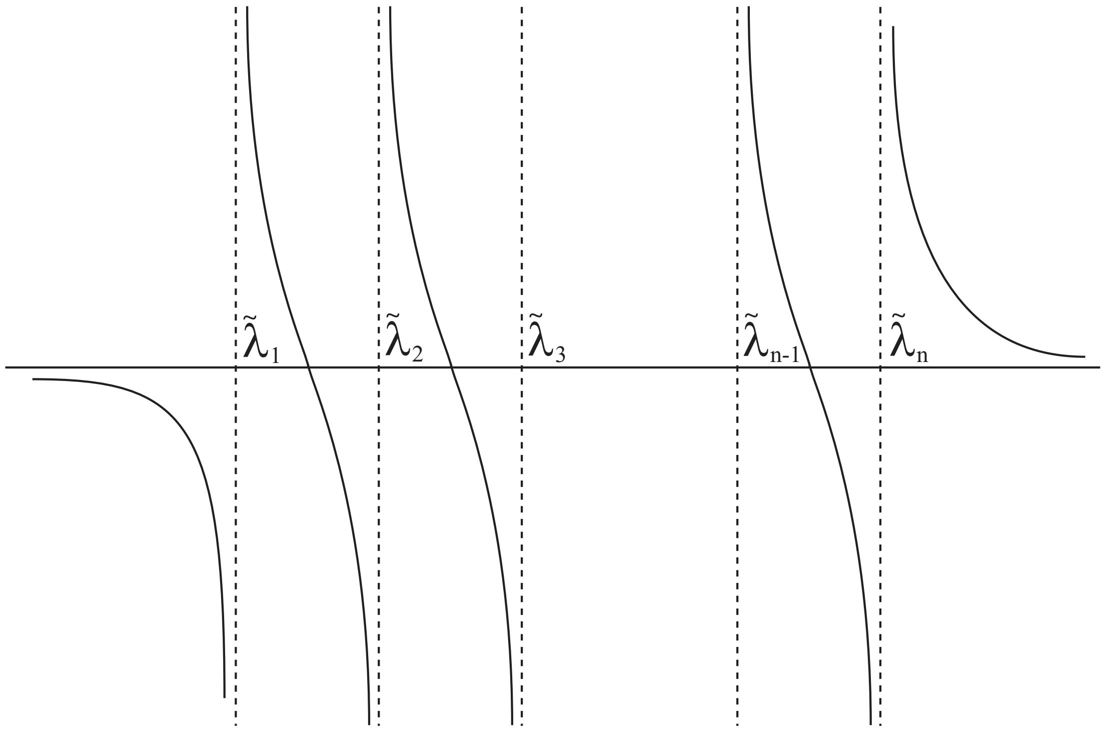

Figure 1.3. Pole and root structure of Stieltjes transforms of a positive measure.

is worth noting that n_h^*/n_l is a probability measure over the set $\{1, \ldots, L\}$, with expectation $\langle\cdot\rangle_L$, say. As $\tilde{m}_l = p_{1,l} n_h^*/n_l$, and $\tilde{\lambda}_l = p_{1,l}$ we may write

$$m_i = \left\langle \frac{p_{1,l}}{(\lambda_i - p_{1,l})^2} \right\rangle_L^{-1}. \tag{1.16}$$

We now verify equation (1.1) for M^{th} order hierarchical series-parallel RRN. By induction, we assume that σ_ω^* satisfies (1.1) for such RRN, for all $M = 0, 1, \ldots, M_0$. We construct $(M_0 + 1)^{\text{th}}$ order of hierarchical series-parallel RRN as follows. First, let $\omega \in \Omega_p$ be a realization of the RRN shown in Figure 1.2 (a) with the σ_l replaced by a M_0^{th} order RRN, $l = 1, \ldots, L$. By induction, the power $\mathcal{H}_\omega$ is given by $\mathcal{H}_\omega = V_0^2 \sigma_{\text{eff}}$, with

$$\frac{\sigma_{\text{eff}}}{\sigma_2} = \frac{1}{\sigma_2} \sum_{l=1}^{L} \frac{\sigma_2}{\tilde{N}_\omega(l)} \left(1 - \sum_{j=1}^{\tilde{L}(l)} \frac{\tilde{m}_j(l)}{s - \tilde{\lambda}_j(l)} \right) = \frac{1}{\tilde{n}_h^*} \left(1 - \sum_{l=1}^{L} \sum_{j=1}^{\tilde{L}(l)} \frac{\tilde{m}_j(l) \tilde{n}_h^* / \tilde{N}_\omega(l)}{s - \tilde{\lambda}_j(l)} \right), \tag{1.17}$$

where $\tilde{n}_h^* = (\sum_{l=1}^{L} \tilde{N}_\omega(l)^{-1})^{-1}$ and $\tilde{n}_h^*/L$ is the harmonic mean of the numbers $\{\tilde{N}_\omega(l)\}$. Define $m_i = \tilde{m}_j(l) \tilde{n}_h^* / \tilde{N}_\omega(l)$ and $\lambda_i = \tilde{\lambda}_j(l)$, where i takes values $i = 1, \ldots, \tilde{L}(l) \times L$, and is defined by $i = i(j, l) = j + (l - 1)\tilde{L}(l)$ for $l = 1, \ldots, L$ and $j = 1, \ldots, \tilde{L}(l)$. With these definitions, (1.17) implies that (1.1) holds with $N_\omega = \tilde{n}_h^*$, $\sigma_\omega^* = \sigma_2(1 - \sum_{i=1}^{L_0} m_i/(s - \lambda_i))$, $L_0 = \tilde{L}(l) \times L$, and $\mu_\omega(d\lambda) = \sum_{i=1}^{L_0} m_i \delta_{\lambda_i}(d\lambda)$. By induction, for each $l = 1, \ldots, L$, $0 \leq \tilde{m}_j(l), \tilde{\lambda}_j(l) \leq 1$, for all $j = 1, \ldots, \tilde{L}(l)$. As $0 < \tilde{n}_h^*/\tilde{N}_\omega(l) \leq 1$, we have $0 \leq \lambda_i, m_i \leq 1$.

Second, let $\omega \in \Omega_p$ be a statistical realization of the RRN shown in Figure 1.2 (b) with the σ_l replaced by a M_0^{th} order hierarchical series-parallel RRN, where $l = 1, \ldots, L$. By induction, the power $\mathcal{H}_\omega$ dissipated by the RRN is given by $\mathcal{H}_\omega = V_0^2 \sigma_{\text{eff}}$, where

$$\begin{aligned}
\frac{\sigma_{\text{eff}}}{\sigma_2} &= \frac{1}{\sigma_2}\left(\sum_{l=1}^{L}\left[\frac{\sigma_2}{\hat{N}_\omega(l)}\left(1 - \sum_{j=1}^{\tilde{L}(l)} \frac{\hat{m}_j(l)}{s - \hat{\lambda}_j(l)}\right)\right]^{-1}\right)^{-1} = \left(\sum_{l=1}^{L} \tilde{N}_\omega(l)(1 - \hat{F}_l(s))^{-1}\right)^{-1} \\
&= \left(\sum_{l=1}^{L} \tilde{N}_\omega(l)\left(1 + \frac{\hat{F}_l(s)}{1 - \hat{F}_l(s)}\right)\right)^{-1} = \left(n_a^*\left(1 + \sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{\hat{F}_l(s)}{1 - \hat{F}_l(s)}\right)\right)^{-1} \\
&= \frac{1}{n_a^*}(1 + \tilde{F}(s))^{-1} = \frac{1}{n_a^*}\left(1 - \frac{\tilde{F}(s)}{1 + \tilde{F}(s)}\right) = \frac{1}{n_a^*}(1 - F(s)) = \frac{1}{n_a^*}\left(1 - \sum_{i=1}^{L_0} \frac{m_i}{s - \lambda_i}\right).
\end{aligned} \tag{1.18}$$

Here we have defined

$$\begin{aligned}
\hat{F}_l(s) &= \sum_{j=1}^{\tilde{L}(l)} \frac{\hat{m}_j(l)}{s - \hat{\lambda}_j(l)}, \quad \tilde{F}(s) = \sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{\hat{F}_l(s)}{1 - \hat{F}_l(s)} = \sum_{l=1}^{L}\sum_{j=1}^{\tilde{L}(l)} \frac{\tilde{m}_j(l)}{s - \tilde{\lambda}_j(l)} = \sum_{i=1}^{L_0} \frac{\tilde{m}_i}{s - \tilde{\lambda}_i} \\
F(s) &= \frac{\tilde{F}(s)}{1 + \tilde{F}(s)} = \sum_{i=1}^{L_0} \frac{m_i}{s - \lambda_i},
\end{aligned} \tag{1.19}$$

and $n_a^* = \sum_{l=1}^{L} \tilde{N}_\omega(l)$, where n_a^*/L is the arithmetic mean of the numbers $\{\tilde{N}_\omega(l)\}$, $i = i(j, l)$ is defined as before, and $L_0 = \tilde{L}(l) \times L$. With these definitions, (1.18) implies that (1.1) holds with $N_\omega = \tilde{n}_a^*$, $\sigma_\omega^* = \sigma_2(1 - \sum_{i=1}^{L_0} m_i/(s - \lambda_i))$, $L_0 = \tilde{L}(l) \times L$, and $\mu_\omega(d\lambda) = \sum_{i=1}^{L_0} m_i \delta_{\lambda_i}(d\lambda)$. We now prove that $0 \leq \lambda_i \leq 1$ and $m_l \geq 0$ for all $i = 1, \ldots, L_0$.

Analogous to equation (1.11), we write $\hat{F}_l(s) = \hat{Q}_{\tilde{L}(l)}(s)/\hat{P}_{\tilde{L}(l)}(s)$, a ratio of polynomials. This implies that $\tilde{F}(s) = Q_{L_0}(s)/P_{L_0}(s)$, a ratio of polynomials, and that $F(s) = Q_{L_0}(s)/(P_{L_0}(s) + Q_{L_0}(s))$. The roots and poles of $\hat{F}_l(s)$ are the roots of the polynomials $\hat{Q}_{\tilde{L}(l)}(s)$ and $\hat{P}_{\tilde{L}(l)}(s)$, respectively, and similarly for $\tilde{F}(s)$ and $F(s)$. As before, without loss of generality, we may assume that the poles of $\tilde{F}(s)$ and $F(s)$ are *distinct* and ordered, e.g. $\lambda_i < \lambda_{i+1}$. By induction, we have $0 \leq \hat{m}_j(l) \leq 1$, for all $l = 1, \ldots, L$ and $j = 1, \ldots, \tilde{L}(l)$. Therefore $\hat{F}_l(s)$ is monotonic decreasing for all $l = 1, \ldots, L$: $\hat{F}_l'(s) = -\sum_{j=1}^{\tilde{L}(l)} \hat{m}_j(l)/(s - \hat{\lambda}_j(l))^2 < 0$. This implies that, for every $l = 1, \ldots, L$, there is precisely one root of $\hat{F}_l(s)$ in the each of the intervals $(\hat{\lambda}_j(l), \hat{\lambda}_{j+1}(l))$, $j = 1, \ldots, \tilde{L}(l) - 1$, so that the roots and poles of $\hat{F}_l(s)$ interlace. Moreover, the functions $\tilde{F}(s)$ and $F(s)$ are also monotonic decreasing:

$$\tilde{F}'(s) = \sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{\hat{F}_l'(s)}{(1-\hat{F}_l(s))^2} < 0, \quad F'(s) = \frac{\tilde{F}_l'(s)}{(1+\tilde{F}_l(s))^2} < 0. \tag{1.20}$$

This implies that the roots and poles of $\tilde{F}(s)$ interlace, and the roots and poles of $F(s)$ interlace. Clearly, the roots of $F(s)$ coincide with the roots of $\tilde{F}(s)$, and the poles λ_i of $F(s)$ are determined by the equation $\tilde{F}(\lambda_i) = -1$. This, implies that the poles of $\tilde{F}(s)$ and $F(s)$ interlace: $\lambda_i < \tilde{\lambda}_i < \lambda_{i+1} < \tilde{\lambda}_{i+1}$, $i = 1, \ldots, L_0 - 1$. Exactly as in (1.14) we have $m_i = -1/\tilde{F}'(\lambda_i)$, and by equation (1.20) we have $\tilde{F}'(s) < 0$, which implies that $m_i > 0$ for all $i = 1, \ldots, L_0$. Furthermore as $\lim_{s \longrightarrow \tilde{\lambda}_i} |\tilde{F}(s)| = \infty$, (1.19) implies that there exist $l \in \{1, \ldots, L\}$ such that $\hat{F}_l(\tilde{\lambda}_i) = 1$. Denoting the set of such l by $\mathcal{L}_\infty$, we have

$$\tilde{m}_i = \sum_{l \in \mathcal{L}_\infty} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{-1}{\hat{F}_l'(\tilde{\lambda}_i)} > 0. \tag{1.21}$$

We now prove that $0 \le \lambda_i \le 1$. By induction, for all $l = 1, \ldots, L$, the poles $\hat{\lambda}_j(l)$ of $\hat{F}_l(s)$ satisfy $0 \le \hat{\lambda}_1(l) < \hat{\lambda}_2(l) < \cdots < \hat{\lambda}_{\tilde{L}(l)}(l) \le 1$. Thus, for all $l = 1, \ldots, L$, and $s < 0$ we have $\hat{F}_l(s) < 0$, and $\hat{F}_l(s) > 0$ for $s > 1$. We first prove that $\lambda_i \ge 0$ for all $i = 1, \ldots, L_0$. Assume the contrary, that the minimum value λ_1 of the $\{\lambda_i\}$ satisfies $\lambda_1 < 0$, which implies that $\hat{F}_l(\lambda_1) < 0$ for all $l = 1, \ldots, L$. Therefore, as the λ_i are determined by the condition $\tilde{F}(\lambda_i) = -1$, we have

$$\begin{aligned} 1 = -\tilde{F}(\lambda_1) &= -\sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{\hat{F}_l(\lambda_1)}{1-\hat{F}_l(\lambda_1)} = \sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{|\hat{F}_l(\lambda_1)|}{1+|\hat{F}_l(\lambda_1)|} \\ &= \sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{1}{1+1/|\hat{F}_l(\lambda_1)|} < \sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} = 1, \end{aligned} \tag{1.22}$$

a contradiction, which implies that $\lambda_i \ge 0$, for all $i = 1, \ldots, L_0$. Similarly, assume that the maximum value λ_{L_0} of the $\{\lambda_i\}$ satisfies $\lambda_{L_0} > 1$, which implies that $\hat{F}_l(\lambda_{L_0}) > 0$ for all $l = 1, \ldots, L$. Therefore,

$$1 = -\tilde{F}(\lambda_{L_0}) = -\sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{\hat{F}_l(\lambda_1)}{1-\hat{F}_l(\lambda_1)} = -\sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{|\hat{F}_l(\lambda_1)|}{1-|\hat{F}_l(\lambda_1)|} \tag{1.23}$$

$$= \sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} \frac{1}{1-1/|\hat{F}_l(\lambda_1)|} > \sum_{l=1}^{L} \frac{\tilde{N}_\omega(l)}{n_a^*} = 1, \tag{1.24}$$

a contradiction, which implies that $\lambda_i \le 1$. In summary, we have $0 \le \lambda_i \le 1$. This concludes our induction step of the proof. Therefore equation (1.1) holds for all M^{th} order hierarchical series-parallel RRN, $M = 0, 1, 2, \ldots$ $\square$.

We now discuss the possibility that $0 \leq m_i \leq 1$ for all M^{th} order hierarchical series-parallel RRN, $M \in \mathbb{Z}^+$. We already proved that $m_i \geq 0$ for all $i = 1, \ldots, L_0$. Recall that the poles $\tilde{\lambda}_i$ and λ_i of $\tilde{F}(s)$ and $F(s)$, respectively, interlace: $0 \leq \lambda_i < \tilde{\lambda}_i < \lambda_{i+1} < \tilde{\lambda}_{i+1}$, $i = 1, \ldots, L_0 - 1$, with $\lambda_{L_0} \leq 1$. If $\tilde{\lambda}_{L_0} < 1$ or if $|\lambda_i - \tilde{\lambda}_j| < 1$ for all $i, j = 1, \ldots, L_0$, then exactly as in (1.15) we have $-\tilde{F}'(\lambda_i) \geq 1$, which implies that $m_i \leq 1$ for all $i = 1, \ldots, L_0$. Clearly $|\lambda_i - \tilde{\lambda}_j| < 1$ for all $i = 1, \ldots, L_0$ and $j = 1, \ldots, L_0 - 1$, but we may have $\tilde{\lambda}_{L_0} > 1$ and $|\lambda_i - \tilde{\lambda}_{L_0}| > 1$ for some $i = 1, \ldots, L_0$. In this case, the following argument provides substantial evidence that $m_i \leq 1$, i.e. $-\tilde{F}'(\lambda_i) \geq 1$, but does not constitute a proof. Denote by $\mathcal{J}_+$ and $\mathcal{J}_-$ the set of $j \in \{1, \ldots, L_0\}$ such that $\lambda_i - \tilde{\lambda}_j > 0$ and $\lambda_i - \tilde{\lambda}_j < 0$, respectively. As $\tilde{F}(\lambda_i) = -1$, we have

$$
\begin{aligned}
-\tilde{F}'(\lambda_i) - 1 &= -\tilde{F}'(\lambda_i) + \tilde{F}(\lambda_i) = \sum_{j=1}^{L_0} \frac{\tilde{m}_j}{(\lambda_i - \tilde{\lambda}_j)^2} + \sum_{j=1}^{L_0} \frac{\tilde{m}_j}{\lambda_i - \tilde{\lambda}_j} \qquad (1.25)\\
&= \sum_{j \in \mathcal{J}_+} \tilde{m}_j \left[\frac{1 + |\lambda_i - \tilde{\lambda}_j|}{|\lambda_i - \tilde{\lambda}_j|^2} \right] + \sum_{L_0 \neq j \in \mathcal{J}_-} \tilde{m}_j \left[\frac{1 - |\lambda_i - \tilde{\lambda}_j|}{|\lambda_i - \tilde{\lambda}_j|^2} \right] + \frac{\tilde{m}_{L_0}}{|\lambda_i - \tilde{\lambda}_j|^2} - \frac{\tilde{m}_{L_0}}{|\lambda_i - \tilde{\lambda}_j|}.
\end{aligned}
$$

We have $|\lambda_i - \tilde{\lambda}_j| < 1$ for all $i = 1, \ldots, L_0$ and $j = 1, \ldots, L_0 - 1$, and by hypothesis $|\lambda_i - \tilde{\lambda}_{L_0}| > 1$. Therefore, only the last term of the second line of (1.25) is negative. Thus, $m_i \leq 1$ only if the magnitude of this last term is less than the sum of all the others. This is very likely but the proof is unclear, given what we have established so far. The details of the condition $m_i \leq 1$, as well as the mass of the measures μ_ω and the infinite number limit $n \longrightarrow \infty$, will be further explored in future work. We now give an analogue of equation (1.16) for $m_i = -1/\tilde{F}'(\lambda_i)$. Note that $\tilde{N}_\omega(l)/n_a^*$ is a probability measure on the set $\{1, \ldots, L\}$ with expectation $\langle \cdot \rangle_L$, say. This analogue thus follows from (1.19):

$$
m_i = \left\langle \frac{-\hat{F}_l'(\lambda_i)}{(1 - \hat{F}_l(\lambda_i))^2} \right\rangle_L^{-1} = \left\langle \frac{\sum_{j=1}^{\tilde{L}(l)} \frac{\hat{m}_j(l)}{(s - \hat{\lambda}_j(l))^2}}{\left(1 - \sum_{j=1}^{\tilde{L}(l)} \frac{\hat{m}_j(l)}{s - \hat{\lambda}_j(l)}\right)^2} \right\rangle_L^{-1}. \qquad (1.26)
$$

We have shown that, for each $\omega \in \Omega_p$, the effective conductance σ_ω^* satisfies equation (1.1) for all M^{th} order hierarchical series-parallel RRN, $M \in \mathbb{Z}^+$. The ensemble averaged effective conductance σ^* may be defined by $\mathcal{H} = \langle \mathcal{H}_\omega \rangle = \tilde{V}_0^2 \sigma^* = \tilde{V}_0^2 \sigma_2 (1 - F(s; \mu))$, where the expectation $\langle \cdot \rangle$ is over Ω_p, $\mathcal{H}$ is the average power dissipated by the RRN, and $\tilde{V}_0$ is the average voltage of the RRN. By equation (1.1) we have

$$\mathcal{H} = \left\langle \frac{V_0^2\sigma_2}{N_\omega}(1 - F(s,\mu_\omega)) \right\rangle = \frac{V_0^2\sigma_2}{\langle N_\omega^{-1}\rangle^{-1}} \left(1 - \left\langle \frac{F(s,\mu_\omega)}{\langle N_\omega^{-1}\rangle N_\omega} \right\rangle \right) = \tilde{V}_0^2\sigma_2(1 - F(s;\mu)),$$

$$F(s;\mu) = \int_0^1 \frac{d\mu(\lambda)}{s-\lambda}, \quad \mu(d\lambda) = \sum_{i=1}^{L_0} \left\langle \frac{m_i(\omega)\delta_{\lambda_i}(d\lambda)}{\langle N_\omega^{-1}\rangle N_\omega} \right\rangle, \quad \tilde{V}_0 = V_0\sqrt{\langle N_\omega^{-1}\rangle} \tag{1.27}$$

A key feature of equations (1.1) and (1.27) is that parameter information in s and V_0 is *separated* from the geometry of the RRN, which is incorporated in the measures μ_ω and μ, respectively. This is a very useful property which will be used extensively in this dissertation.

In [65], Golden and Papanicolaou formulated an abstract theory was given which yields Stieltjes integral representations of σ^* for general two-component stationary random media in lattice and continuum settings. This work reveals that the theoretical foundations of the theory is in functional analysis, and demonstrates that μ is the spectral measure of a self-adjoint random operator (or matrix). In [58], Golden explicitly formulated this abstract framework for RRN with arbitrary graph topology and for the bond lattice. The explicitness of our construction of equation (1.27) illuminates many key features of the general abstract theory, discussed in Section 2.1, and introduces methods of Padé approximation and the theory of polynomials orthogonal to the measure μ, discussed in Chapter 4. In Section 1.2 we analyze a continuum analogue of the RRN shown in Figure 1.2.

1.2 Laminate Dielectric Media

This section is devoted to an analysis of two-phase random laminate media. These simple continuum microstructures are central to the theory of two-phase composites as they are, in a limiting sense, the building blocks of all such composite media [87]. These canonical examples of two-phase random media are analogous to the parallel and series RRN shown in Figure 1.2 of Section 1.1.

In continuum composite media, the electric field $\vec{E}(\vec{x},\omega)$ plays the role of the voltage V_j across resistor j of a RRN, while the random geometry $\omega \in \Omega$ of a continuum composite plays the role of the random configuration of resistors of a RRN, where $\vec{x} \in \mathbb{R}^d$, d is the physical dimension of the medium, and Ω is the set of all geometric realizations of the random medium. Here we demonstrate that an analogue of the parameter separation property, $\mathcal{H} = \tilde{V}_0^2\sigma_2(1 - F(s;\mu))$, displayed in (1.27), holds for two-phase laminate media, where the norm E_0 of the average electric field $\vec{E}_0 = \langle\vec{E}\rangle$ plays the role of $\tilde{V}_0$ in RRN. While this parameter separation property holds for general two-component stationary random media in lattice and continuum settings [65], for laminate geometry, we prove that the condition

$\langle\vec{E}\rangle = \vec{E}_0$ is necessary and sufficient for this property.

We now briefly review the effective parameter problem for two-phase random dielectric media. A more detailed discussion of the effective parameter problem will be given in Section 2.1. Let (Ω, P) be a probability space and let $\epsilon(\vec{x}, \omega)$ be the local permittivity, where Ω is the set of all realizations of our random medium and $P(d\omega)$ is the underlying probability measure [65]. We assume $\epsilon(\vec{x}, \omega)$ takes the values ϵ_1 and ϵ_2 and write $\epsilon(\vec{x}, \omega) = \epsilon_1\chi_1(\vec{x}, \omega) + \epsilon_2\chi_2(\vec{x}, \omega)$, where χ_j is the characteristic function of medium, $j = 1, 2$, which equals one for all $\omega \in \Omega$ having medium j at $\vec{x}$, and zero otherwise [65]. Let $\vec{E}(\vec{x})$ and $\vec{D}(\vec{x})$ be the random electric and displacement fields, related by $\vec{D} = \epsilon\vec{E}$, satisfying [65]

$$\vec{\nabla} \times \vec{E} = 0, \quad \vec{\nabla} \cdot \vec{D} = 0, \qquad \vec{E} = \vec{E}_0 + \vec{E}_f, \quad \langle\vec{E}\rangle = \vec{E}_0, \tag{1.28}$$

where $\langle\cdot\rangle$ denotes averaging and $\vec{E}_f$ is the fluctuating field with mean zero about $\vec{E}_0$. We write $\vec{E}_0 = E_0\vec{e}_0$, where $\vec{e}_0$ is a unit vector which gives the direction of $\vec{E}_0$. For simplicity, we have assumed that the constituents are ideal (perfect electrical insulators [102]), so that $\vec{\nabla} \cdot \vec{D} = \rho_f = 0$, and linear, so that the bound charge distribution ρ_b is directly proportional to the free charge density ρ_f, $\rho_b \propto \rho_f = 0$ [103]. Therefore, the system may be thought of as free space partitioned by the boundaries of the constituents, which may host bound surface charge densities σ_b, where σ is not to be confused with conductance.

The effective permittivity tensor $\boldsymbol{\epsilon}^*$ is defined as

$$\langle\vec{D}\rangle = \boldsymbol{\epsilon}^*\langle\vec{E}\rangle. \tag{1.29}$$

Central to our studies is the system energy given by $\frac{1}{2}\,\langle\vec{D}\cdot\vec{E}\rangle$ [74]. A key variational calculation [65] yields the energy constraint $\langle\vec{D}\cdot\vec{E}_f\rangle = 0$. Therefore the energy may be expressed as

$$\frac{1}{2}\,\langle\vec{D}\cdot\vec{E}\rangle = \frac{1}{2}\,\langle\vec{D}\rangle\cdot\vec{E}_0 = \frac{1}{2}\,\epsilon^* E_0^2, \tag{1.30}$$

where $\epsilon^* = \boldsymbol{\epsilon}^*\vec{e}_0\cdot\vec{e}_0$. For laminates, we will show that the energy constraint $\langle\vec{D}\cdot\vec{E}_f\rangle = 0$ is a direct consequence of the property $\langle\vec{E}\rangle = \vec{E}_0$ and boundary conditions, where $\langle\cdot\rangle$ will denote volume average over all $\mathbb{R}^d$ for the remainder of this section.

As the results of this section are valid for multicomponent media, we will use the notation $\langle\chi_j\rangle = p_j$ for the characteristic function χ_j and volume fraction p_j of material component $j = 1, \ldots, n$. By linearity of the material, we have the following identities

$$\epsilon_0\vec{E} = \vec{D} - \vec{P}_0 = \epsilon\vec{E} - (\epsilon - \epsilon_0)\vec{E} \iff \epsilon_2\vec{E} = \vec{D} - \vec{P}_2 = \epsilon\vec{E} - (\epsilon - \epsilon_2)\vec{E}, \tag{1.31}$$

where $\vec{P}$ is the polarization density. Therefore ϵ_2 can be used in place of the permittivity of free space ϵ_0 in the definition $\epsilon_0 \vec{\nabla} \cdot \vec{E} = \rho_T$ of the total charge density $\rho_T = \rho_f + \rho_b$ [66, 74], without physical nor mathematical inconsistencies.

1.2.1 Laminates Parallel to an Applied Field

Consider a n phase dielectric random medium filling all of $\mathbb{R}^d$ with laminate geometry parallel to a uniform, applied electric field $\vec{E}$, as shown in Figure 1.4 (a) for $n = 2$. This composite geometry is analogous to the network of parallel resistors shown in Figure 1.2. The effective permittivity ϵ^* of this geometry is known [110] to be given by $\epsilon^* = \epsilon_a^*$, where $\epsilon_a^* = \sum_{j=1}^n p_j \epsilon_j$ is the arithmetic mean of the $\{\epsilon_j\}$, $\epsilon_a^* = \epsilon_2(1 - p_1/s)$ when $n = 2$, and $s = 1/(1 - \epsilon_1/\epsilon_2)$. We will derive this formula for ϵ^* through energetic considerations.

Let $\omega \in \Omega$ be a random configuration of a such a n phase dielectric medium. The electric field is curl free $\vec{\nabla} \times \vec{E} = 0$. This causes its tangential component to be continuous across the constituent boundaries [74], $(\vec{E}_j - \vec{E}_{j+1}) \times \vec{n} = 0$, so that it remains parallel to these boundaries throughout the system, and constant in each of the constituents, where $\vec{n}$ is a unit vector, perpendicular to these boundaries and $\vec{E}(\vec{x}) = \vec{E}_j$ when $\epsilon(\vec{x}) = \epsilon_j$. This and symmetry implies that $\vec{E}_j = \vec{E}_{j+1}$ for all j which, in turn implies that $\vec{E}_j = \vec{E}_i$ for all i, j, and that no surface charge densities are induced on the contrast boundaries [74]: $\sigma_{j,j+1} = \epsilon_2(\vec{E}_j - \vec{E}_{j+1}) \cdot \vec{n} = 0$. The condition $\langle \vec{E} \rangle = \vec{E}_0$ now implies that $\vec{E}_j = \vec{E}_0$ for all j, or $\vec{E} \equiv \vec{E}_0$. As $\vec{E} \equiv \vec{E}_0$, $\vec{D} = \epsilon \vec{E}$, and $\epsilon = \sum_{j=1}^n \epsilon_j \chi_j$, the energy $\mathcal{H}$ is given by

$$\mathcal{H} = \left\langle \frac{1}{2} \vec{D} \cdot \vec{E} \right\rangle = \left\langle \frac{1}{2} \left(\sum_{j=1}^n \epsilon_j \chi_j \right) E_0^2 \right\rangle = \frac{1}{2} \epsilon_a^* E_0^2 = \frac{1}{2} \epsilon_2 E_0^2 \left(1 - \frac{p_1}{s} \right), \tag{1.32}$$

where the last equality holds only for two-component composite media. Like its RRN analogue, ϵ_a^* is independent of the configuration $\omega \in \Omega$ of the laminate.

We stress that it was boundary conditions which caused $\vec{E}$ to be constant. In this case of laminate geometry parallel to an applied field, the conditions $\langle \vec{E} \rangle = \vec{E}_0$ and $\langle \vec{E}_f \rangle = 0$, where $\vec{E}_f = \vec{E} - \langle \vec{E} \rangle$, are trivial definitions of the vectors $\vec{E}_0$ and $\vec{E}_f$. Consequently, the energy $\mathcal{H}$ in (1.32) is trivially given by $\mathcal{H} = \frac{1}{2} \langle \vec{D} \rangle \cdot \vec{E}_0$. The converse, if $\langle \vec{D} \cdot \vec{E}_f \rangle = 0$ then $\langle \vec{E}_f \rangle = 0$, is equally trivial in this case. However, we demonstrate in Section 1.2.2 that, in the case of laminate geometry *perpendicular* to an applied field, this nontrivial equivalence continues to hold.

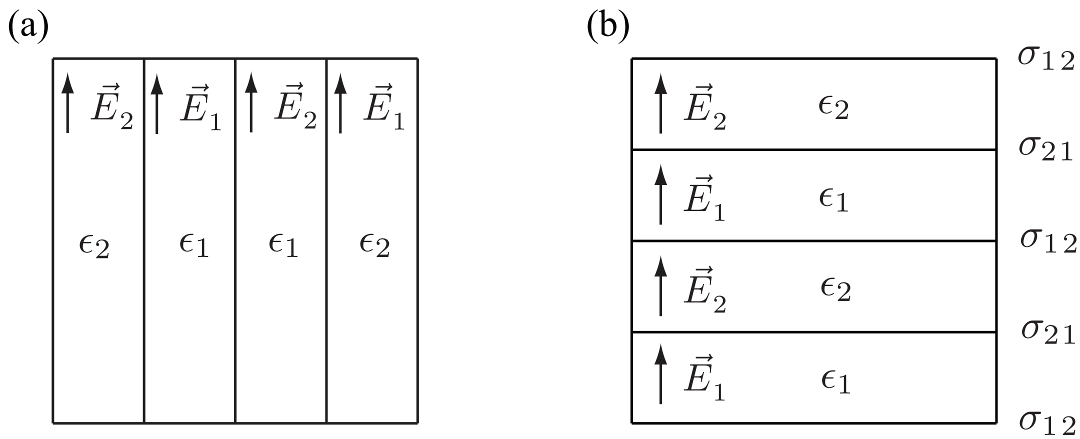

Figure 1.4. (a) Laminates parallel to an applied electric field (b) Laminates perpendicular to an applied electric field

1.2.2 Laminates Perpendicular to an Applied Field

Consider a n phase dielectric random medium filling all of $\mathbb{R}^d$ with laminate geometry perpendicular to a uniform, applied electric field $\vec{E}$, as shown in Figure 1.4 (b) for $n = 2$. This composite geometry is analogous to the network of series resistors shown in Figure 1.2. The effective permittivity ϵ^* of this geometry is known [110] to be given by $\epsilon^* = \epsilon_h^*$, where $\epsilon_h^* = (\sum_{j=1}^n p_j/\epsilon_j)^{-1}$ is the harmonic mean of the $\{\epsilon_j\}$ and $\epsilon_h^* = \epsilon_2(1 - p_1/(s - p_2))$ when $n = 2$. We will derive this formula for ϵ^* through energetic considerations and explore its consequences, which will illuminate features of the effective permittivity ϵ^* for general two-component stationary random media, discussed in Section 2.1.

Let $\omega \in \Omega$ be a random configuration of such a n phase dielectric medium. As the dielectric constituents are ideal, the free charge density is zero and the displacement field is divergence free $\vec{\nabla} \cdot \vec{D} = \rho_f = 0$. This causes its normal component to be continuous across the constituent boundaries [74], $(\epsilon_j \vec{E}_j - \epsilon_{j+1}\vec{E}_{j+1}) \cdot \vec{n} = 0$, so that the electric field $\vec{E}$ remains perpendicular to these boundaries throughout the system and the displacement field $\vec{D}$ is constant in each of the laminate layers. Here $\vec{n} = \vec{e}_0$ is the unit normal to the contrast boundaries and $\vec{E}(\vec{x}) = \vec{E}_j = E_j \vec{e}_0$ when $\epsilon(\vec{x}) = \epsilon_j$. This and symmetry implies that $\epsilon_j \vec{E}_j = \epsilon_{j+1}\vec{E}_{j+1}$ for all j which, in turn implies that $\epsilon_j \vec{E}_j = \epsilon_i \vec{E}_i$ for all i, j.

By the above analysis, the local permittivity ϵ and electric field $\vec{E}$ may be written as $\epsilon = \sum_{j=1}^n \epsilon_j \chi_j$ and $\vec{E} = \sum_{j=1}^n \chi_j \vec{E}_j$, respectively. Focusing on $\vec{E}_1$, we have $\epsilon_j \vec{E}_j = \epsilon_1 \vec{E}_1$ for all j. The condition $\langle \vec{E} \rangle = \vec{E}_0$ then implies that

$$\vec{E}_0 = \sum_{j=1}^{n} p_j \vec{E}_j = \epsilon_1 \vec{E}_1 \sum_{j=1}^{n} \frac{p_j}{\epsilon_j} = \frac{\epsilon_1}{\epsilon_h^*} \vec{E}_1. \tag{1.33}$$

Equation (1.33) yields a global continuity equation for the displacement field:

$$\epsilon_j \vec{E}_j = \epsilon_h^* \vec{E}_0, \quad j = 1, 2, \ldots, n. \tag{1.34}$$

By (1.34) and the orthogonality of the χ_i, $\chi_i \chi_j = \delta_{ij} \chi_j$, the system energy $\mathcal{H}$ is given by

$$\begin{aligned} \mathcal{H} &= \frac{1}{2}\langle \vec{D} \cdot \vec{E} \rangle = \frac{1}{2}\langle \epsilon \vec{E} \cdot \vec{E} \rangle = \frac{1}{2}\left\langle \left(\sum_{j=1}^{n} \epsilon_j \chi_j\right) \left(\sum_{j=1}^{n} \chi_j \vec{E}_j\right) \cdot \left(\sum_{j=1}^{n} \chi_j \vec{E}_j\right) \right\rangle \\ &= \frac{1}{2}\sum_{j=1}^{n} p_j \epsilon_j E_j^2 = \frac{1}{2}\sum_{j=1}^{n} p_j \epsilon_j \left(\frac{\epsilon_h^* E_0}{\epsilon_j}\right)^2 \\ &= \frac{1}{2}\epsilon_h^* E_0^2 = \frac{1}{2}\epsilon_2 E_0^2 \left(1 - \frac{p_1}{s - p_2}\right), \end{aligned} \tag{1.35}$$

where the last equality holds only for two-component media. Like its RRN analogue, ϵ_a^* is independent of the configuration $\omega \in \Omega$ of the laminate. By (1.34) the surface charge densities induced on the constituent boundaries are given by [74]

$$\begin{aligned} \sigma_{i,i+1} &= \epsilon_2(\vec{E}_i - \vec{E}_{i+1}) \cdot \vec{n} = E_0 \epsilon_2 \epsilon_h^* \left(\frac{1}{\epsilon_i} - \frac{1}{\epsilon_{i+1}}\right) \\ &= \pm E_0 \epsilon_2 \epsilon_h^* \left(\frac{1}{\epsilon_1} - \frac{1}{\epsilon_2}\right) = \pm E_0 \epsilon_h^* \frac{1-h}{h} = \pm \frac{E_0 \epsilon_h^*}{s-1}, \end{aligned} \tag{1.36}$$

where the equalities in the second line of equation (1.36) hold only for two-component composite media, and we have used $h = \epsilon_1/\epsilon_2 = (s-1)/s$.

The following theorem illustrates that, for this special geometry, the energy constraint $\langle \vec{D} \cdot \vec{E}_f \rangle = 0$, the condition $\langle \vec{E} \rangle = \vec{E}_0$, and equation (1.34) are equivalent statements.

Theorem 2 *Consider a n phase dielectric random medium filling all of* $\mathbb{R}^d$ *with laminate geometry perpendicular to a uniform, applied electric field* $\vec{E}$, *with* $\vec{E} = \vec{E}_0 + \vec{E}_f$, *where* $\vec{E}_f$ *is the fluctuating field of mean zero about its average* $\langle \vec{E} \rangle = \vec{E}_0$, *and* $\langle \cdot \rangle$ *denotes volume averaging. Let* $\vec{D} = \epsilon \vec{E}$ *be the displacement field, where* ϵ *is the local permittivity of the medium. Let the constituent permittivities* ϵ_j *take volume fractions* p_j *and define* $\epsilon_h^* = (\sum_{j=1}^{n} p_j/\epsilon_j)^{-1}$. *Then the following statements are equivalent:*

(1) $\langle \vec{E}_f \rangle = 0$, ***(2)*** $\epsilon_j \vec{E}_j = \epsilon_h^* \vec{E}_0, \quad j = 1, 2, \ldots, n$, ***(3)*** $\langle \vec{D} \cdot \vec{E}_f \rangle = 0$.

Proof: The proof of **(1)⇒(2)** has already been established by equation (1.34). Conversely, **(1)⇐(2),** if $\epsilon_j \vec{E}_j = \epsilon_h^* \vec{E}_0$ for $j = 1, 2, \ldots, n$ then

$$\langle \vec{E}_f \rangle = \left\langle \sum_{j=1}^{n} \chi_j \vec{E}_j \right\rangle - \vec{E}_0 = \vec{E}_0 \left(\epsilon_h^* \sum_{j=1}^{n} \frac{p_j}{\epsilon_j} - 1 \right) = 0.$$

The property **(2)⇒(3)** follows from the definition $\vec{E} = \vec{E}_f + \vec{E}_0$, the orthogonality of the χ_j, $\chi_i \chi_j = \delta_{ij} \chi_j$, and the symmetry of the problem, which yields $\vec{E}_j = E_j \vec{e}_0$, for all $j = 0, 1, \ldots, n$, and $\vec{E}_0 = E_0 \vec{e}_0$. Indeed, if $\epsilon_j \vec{E}_j = \epsilon_h^* \vec{E}_0$ for all $j = 1, 2, \ldots, n$, then

$$\begin{aligned} \langle \vec{D} \cdot \vec{E}_f \rangle = \langle \epsilon \vec{E} \cdot \vec{E}_f \rangle &= \left\langle \left(\sum_{j=1}^{n} \epsilon_j \chi_j \right) \left(\sum_{j=1}^{n} \chi_j \vec{E}_j \right) \cdot \left(\sum_{j=1}^{n} \chi_j \vec{E}_j - \vec{E}_0 \right) \right\rangle \\ &= \left\langle \sum_{j=1}^{n} \chi_j (\epsilon_j E_j^2 - \epsilon_j E_j E_0) \right\rangle = \sum_{j=1}^{n} p_j \left(\epsilon_j \left(\frac{\epsilon_h^*}{\epsilon_j} \right)^2 - \epsilon_j \frac{\epsilon_h^*}{\epsilon_j} \right) E_0^2 = 0, \end{aligned} \tag{1.37}$$

where we have used $\sum_{j=1}^{n} p_j = 1$ in the last line. Conversely **(2)⇐(3)**, the electric field definition, the symmetry of the problem, and the orthogonality of the χ_j implies all but the last two equalities of equation (1.37). Therefore if $\langle \vec{D} \cdot \vec{E}_f \rangle = 0$ then the boundary condition, $\epsilon_j E_j = \epsilon_1 E_1$ for $j = 1, 2, \ldots, n$, yields

$$\begin{aligned} 0 = \langle \vec{D} \cdot \vec{E}_f \rangle &= \sum_{j=1}^{n} p_j (\epsilon_j E_j^2 - \epsilon_j E_j E_0) = \sum_{j=1}^{n} p_j \left(\epsilon_j \left(\frac{\epsilon_1 E_1}{\epsilon_j} \right)^2 - \epsilon_j \frac{\epsilon_1 E_1}{\epsilon_j} E_0 \right) \\ &= \epsilon_1 E_1 \left(\frac{\epsilon_1 E_1}{\epsilon_h^*} - E_0 \right). \end{aligned}$$

This and the boundary condition then implies that $\epsilon_j \vec{E}_j = \epsilon_h^* \vec{E}_0$ for all $j = 1, 2, \ldots, n$. The equivalence **(1)** $\Longleftrightarrow$ **(3)** is therefore also established. This concludes the proof of Theorem 2 □.

In Theorem 3 we provide a detailed decomposition of the system energy.

Theorem 3 *Consider the dielectric random medium described in Theorem 2. In addition to the definitions given there, denote by* $\epsilon_a^* = \sum_{j=1}^{n} \epsilon_j p_j$ *the arithmetic mean of the constituent permittivities* $\{\epsilon_j\}$*. Then we have the following decomposition of the system energy* $\frac{1}{2} \langle \vec{D} \cdot \vec{E} \rangle = \frac{1}{2} \langle \epsilon \vec{E} \cdot \vec{E} \rangle$*:*

$$\begin{gathered} \langle \epsilon \vec{E} \cdot \vec{E} \rangle = E_0^2 \epsilon_h^*, \quad \langle \epsilon \vec{E} \cdot \vec{E}_f \rangle = 0, \quad \langle \epsilon \vec{E} \cdot \vec{E}_0 \rangle = E_0^2 \epsilon_h^*, \\ \langle \epsilon \vec{E}_f \cdot \vec{E}_0 \rangle = E_0^2 (\epsilon_h^* - \epsilon_a^*), \quad \langle \epsilon \vec{E}_f \cdot \vec{E}_f \rangle = E_0^2 (\epsilon_a^* - \epsilon_h^*), \end{gathered}$$

Proof: The formula $\langle \epsilon \vec{E} \cdot \vec{E} \rangle = E_0^2 \epsilon_h^*$ has already been established in equation (1.35). By Theorem 2, $\langle \epsilon \vec{E} \cdot \vec{E}_f \rangle = 0$, therefore $\vec{E} = \vec{E}_0 + \vec{E}_f$ implies that $\langle \epsilon \vec{E} \cdot \vec{E}_0 \rangle = E_0^2 \epsilon_h^*$. By the orthogonality of the χ_j, $\chi_i \chi_j = \delta_{ij} \chi_j$, the definition $\vec{E} = \vec{E}_0 + \vec{E}_f$, equation (1.34), and the symmetry of the problem, which yields $\vec{E}_j = E_j \vec{e}_k$, for all $j = 0, 1, \ldots, n$, and $\vec{E}_0 = E_0 \vec{e}_k$, we have

$$\begin{aligned} \langle \epsilon \vec{E}_f \cdot \vec{E}_0 \rangle &= \left\langle \left(\sum_{j=1}^{n} \epsilon_j \chi_j \right) \left(\sum_{j=1}^{n} \chi_j \vec{E}_j - \vec{E}_0 \right) \cdot \vec{E}_0 \right\rangle = \left\langle \sum_{j=1}^{n} \epsilon_j \chi_j (E_j - E_0) E_0 \right\rangle \\ &= E_0^2 \sum_{j=1}^{n} p_j \epsilon_j \left(\frac{\epsilon_h^*}{\epsilon_j} - 1 \right) = E_0^2 (\epsilon_h^* - \epsilon_a^*), \end{aligned}$$

where we have used $\sum_{j=1}^{n} p_j = 1$. By $\langle \epsilon \vec{E} \cdot \vec{E}_f \rangle = 0$ and $\vec{E} = \vec{E}_0 + \vec{E}_f$ we therefore have $\langle \epsilon \vec{E}_f \cdot \vec{E}_f \rangle = E_0^2 (\epsilon_a^* - \epsilon_h^*)$. This concludes the proof of Theorem 3 $\square$.

In Theorem 4 we give the analogue of this result for general two-component stationary random media. This theorem makes many results of this dissertation physically transparent. We will use the intuition gained from Section 1.1 and Section 1.2 to guide us in our exploration of the general features of two-phase random media, in lattice and continuum settings.

CHAPTER 2

EFFECTIVE PARAMETERS OF TWO-PHASE RANDOM MEDIA

We now formulate the effective parameter problem for general two-phase stationary conductive media, in lattice and continuum settings. By the symmetries in the equations governing two-phase dielectric and conductive media [87], the results given here also hold for binary dielectric composites. The mathematical objects arising in this chapter lead to connections with percolation theory, statistical mechanics, orthogonal polynomial theory, and random matrix theory.

2.1 The Analytic Continuation Method

Let (Ω, P) be a probability space, and let $\boldsymbol{\sigma}(\vec{x},\omega)$ and $\boldsymbol{\rho}(\vec{x},\omega)$ be the local conductivity and resistivity tensors, respectively, which are (spatially) stationary random fields in $\vec{x} \in \mathbb{R}^d$ and $\omega \in \Omega$. Here Ω is the set of all geometric realizations of our random medium, $P(d\omega)$ is the underlying probability measure, which is compatible with stationarity, and $\boldsymbol{\rho} = \boldsymbol{\sigma}^{-1}$ [65]. Define the Hilbert space of stationary random fields $\mathscr{H}_s \subset L^2(\Omega, P)$, and the underlying Hilbert spaces of stationary curl free $\mathscr{H}_\times \subset \mathscr{H}_s$ and divergence free $\mathscr{H}_\bullet \subset \mathscr{H}_s$ random fields

$$\mathscr{H}_\times = \{\vec{Y}(\omega) \in \mathscr{H}_s \mid \vec{\nabla} \times \vec{Y} = 0 \text{ weakly and } \langle \vec{Y} \rangle = 0\}, \tag{2.1}$$
$$\mathscr{H}_\bullet = \{\vec{Y}(\omega) \in \mathscr{H}_s \mid \vec{\nabla} \cdot \vec{Y} = 0 \text{ weakly and } \langle \vec{Y} \rangle = 0\},$$

where $\vec{Y} : \Omega \mapsto \mathbb{R}^d$ and $\langle \cdot \rangle$ means ensemble average over Ω, or by an ergodic theorem spatial average over all of $\mathbb{R}^d$ [65].

Consider the variational problems [65]: find $\vec{E}_f \in \mathscr{H}_\times$ and $\vec{J}_f \in \mathscr{H}_\bullet$ such that

$$\langle \boldsymbol{\sigma}(\vec{E}_0 + \vec{E}_f) \cdot \vec{Y} \rangle = 0 \quad \forall\, \vec{Y} \in \mathscr{H}_\times \qquad \text{and} \qquad \langle \boldsymbol{\rho}(\vec{J}_0 + \vec{J}_f) \cdot \vec{Y} \rangle = 0 \quad \forall\, \vec{Y} \in \mathscr{H}_\bullet\,, \tag{2.2}$$

respectively. When the bilinear forms $a(\vec{u},\vec{v}) = \vec{u}^T \boldsymbol{\sigma}\, \vec{v}$ and $\tilde{a}(\vec{u},\vec{v}) = \vec{u}^T \boldsymbol{\rho}\, \vec{v}$ are bounded and coercive, these problems have unique solutions satisfying [65]

$$\vec{\nabla} \times \vec{E} = 0, \qquad \vec{\nabla} \cdot \vec{J} = 0, \qquad \vec{J} = \boldsymbol{\sigma} \vec{E}, \qquad \vec{E} = \vec{E}_0 + \vec{E}_f, \qquad \langle \vec{E}\,\rangle = \vec{E}_0, \tag{2.3}$$
$$\vec{\nabla} \times \vec{E} = 0, \qquad \vec{\nabla} \cdot \vec{J} = 0, \qquad \vec{E} = \boldsymbol{\rho} \vec{J}, \qquad \vec{J} = \vec{J}_0 + \vec{J}_f, \qquad \langle \vec{J}\,\rangle = \vec{J}_0,$$

respectively. Here $\vec{E}_f$ and $\vec{J}_f$ are the fluctuating electric field and current density of mean zero, respectively, about the (constant) averages $\vec{E}_0$ and $\vec{J}_0$, respectively.

We assume that the local conductivity $\sigma(\vec{x}, \omega)$ of the medium takes the *complex* values σ_1 and σ_2 and write $\sigma(\vec{x}, \omega) = \sigma_1 \chi_1(\vec{x}, \omega) + \sigma_2 \chi_2(\vec{x}, \omega)$, where χ_j is the characteristic function of medium $j = 1, 2$, which equals one for all $\omega \in \Omega$ having medium j at $\vec{x}$, and zero otherwise, with $\chi_1 = 1 - \chi_2$ [65]. Similarly, we assume that the local resistivity $\rho(\vec{x}, \omega)$ takes the values $1/\sigma_1$ and $1/\sigma_2$ and write $\rho(\vec{x}, \omega) = \chi_1(\vec{x}, \omega)/\sigma_1 + \chi_2(\vec{x}, \omega)/\sigma_2$.

As $\vec{E}_f \in \mathscr{H}_\times$ and $\vec{J}_f \in \mathscr{H}_\bullet$, equation (2.2) yields the energy (power density) constraints $\langle \vec{J} \cdot \vec{E}_f \rangle = \langle \vec{E} \cdot \vec{J}_f \rangle = 0$, which lead to the reduced energy representations

$$\langle \vec{J} \cdot \vec{E} \rangle = \langle \vec{J} \rangle \cdot \vec{E}_0 \quad \text{and} \quad \langle \vec{E} \cdot \vec{J} \rangle = \langle \vec{E} \rangle \cdot \vec{J}_0\,. \tag{2.4}$$

The effective complex conductivity and resistivity tensors, $\boldsymbol{\sigma}^*$ and $\boldsymbol{\rho}^*$, are defined by

$$\langle \vec{J}\,\rangle = \boldsymbol{\sigma}^* \vec{E}_0 \quad \text{and} \quad \langle \vec{E}\,\rangle = \boldsymbol{\rho}^* \vec{J}_0, \tag{2.5}$$

respectively, yielding $\langle \vec{J} \cdot \vec{E} \rangle = \boldsymbol{\sigma}^* \vec{E}_0 \cdot \vec{E}_0 = \boldsymbol{\rho}^* \vec{J}_0 \cdot \vec{J}_0$. For simplicity we focus on one diagonal component of these tensors, $\sigma^* = \sigma^*_{kk}$ and $\rho^* = \rho^*_{kk}$, for some $k = 1, \ldots, d$. Assuming that $0 < |\sigma_1| < |\sigma_2| < \infty$, these functions have the following bounds [87, 122]

$$|\sigma_1\,| \leq |\sigma^*| \leq |\sigma_2|, \qquad |\sigma_2|^{-1} \leq |\rho^*| \leq |\sigma_1|^{-1}. \tag{2.6}$$

Due to the homogeneity of these functions, e.g. $\sigma^*(a\sigma_1, a\sigma_2) = a\sigma^*(\sigma_1, \sigma_2)$ for any complex number a, they depend only on the ratio $h = \sigma_1/\sigma_2$, and we define the functions

$$m(h) = \sigma^*/\sigma_2, \quad w(z) = \sigma^*/\sigma_1, \quad \tilde{m}(h) = \sigma_1 \rho^*, \quad \tilde{w}(z) = \sigma_2 \rho^*, \tag{2.7}$$

where $z = 1/h$. The dimensionless functions $m(h)$ and $\tilde{m}(h)$ are analytic off the negative real axis in the h-plane, while $w(z)$ and $\tilde{w}(z)$ are analytic off the negative real axis in the z-plane [65]. Each take the corresponding upper half plane to the upper half plane, so that they are examples of Herglotz functions [65]. As a function of h, $z : (-\infty, 0) \mapsto (-\infty, 0)$. Therefore the functions $w(z(h))$ and $\tilde{w}(z(h))$ are also analytic off the negative real axis in the h-plane. We henceforth restrict h in the complex plane to the set

$$\mathcal{U}_\varepsilon = \{h \in \mathbb{C} : |h| < 1 \text{ and } |h - h_0| > \varepsilon \text{ for all } h_0 \in (-1, 0]\}, \tag{2.8}$$

which is parameterized by $0 < \varepsilon \ll 1$. When $\varepsilon = 0$ in equation (2.8) we write $\mathcal{U}_0$.

A key step in the method is obtaining integral representations for σ^* and ρ^* in terms of Herglotz functions $\mathcal{A}_{i,j}$ and $\mathcal{S}_{i,j}$, $i,j = 0,1,2,\ldots$, of the form [71]

$$\mathcal{A}_{i,j}(\xi;\nu) = \int_0^1 \frac{\lambda^i d\nu(\lambda)}{(\xi-\lambda)^j}\,, \quad \mathcal{S}_{i,j}(\xi;\nu) = \int_0^\infty \frac{y^i d\nu(y)}{(1+\xi y)^j}\,, \tag{2.9}$$

which follow from resolvent representations of the electric field $\vec{E}$ and current density $\vec{J}$,

$$\vec{E} = s(s-\Gamma\chi_1)^{-1}\vec{E}_0 = t(t-\Gamma\chi_2)^{-1}\vec{E}_0 \quad \text{and} \quad \vec{J} = s(s-\Upsilon\chi_2)^{-1}\vec{J}_0 = t(t-\Upsilon\chi_1)^{-1}\vec{J}_0, \tag{2.10}$$

respectively. Here we have defined $s = 1/(1-h)$, $t = 1/(1-z) = 1-s$, $\Gamma = \vec{\nabla}\Delta^{-1}\vec{\nabla}\cdot$, and $\Upsilon = -\vec{\nabla}\times\Delta^{-1}\vec{\nabla}\times$, where $\Delta = \nabla^2$ is the Laplacian. These formulas follow from manipulations of equation (2.3), which will be discussed in more detail below.

The operator Γ is a projection onto curl-free fields, based on convolution with the free-space Green's function for the Laplacian [65]. More specifically $\Gamma : \mathscr{H}_s \mapsto \mathscr{H}_\times$, and for every $\vec{\zeta} \in \mathscr{H}_\times$ we have $\Gamma\vec{\zeta} = \vec{\zeta}$. For the convenience of the reader we recall a few vector calculus facts. For every $\vec{\zeta} \in \mathscr{H}_\bullet$ we have $\vec{\zeta} = \vec{\nabla}\times(\vec{A}+\vec{C})$ weakly, where $\vec{\nabla}\times\vec{C} = 0$ weakly [54, 74]. The arbitrary vector $\vec{C}$ can be chosen so that the vector potential $\vec{A}$ satisfies $\vec{\nabla}\cdot\vec{A} = 0$ weakly [74]. Hence, $\vec{\nabla}\times\vec{\zeta} = \vec{\nabla}\times\vec{\nabla}\times\vec{A} = \vec{\nabla}(\vec{\nabla}\cdot\vec{A}) - \Delta\vec{A} = -\Delta\vec{A}$ weakly. The vector $\vec{C}$ chosen in this manner gives the Coulomb (or transverse) *gauge* of $\vec{\zeta}$ [74]. Choosing the members of the Hilbert space $\mathscr{H}_\bullet$ to have Coulomb gauge, one can similarly show that the operator Υ is a projection onto divergence-free fields. More specifically $\Upsilon : \mathscr{H}_s \mapsto \mathscr{H}_\bullet$, and for every $\vec{\zeta} \in \mathscr{H}_\bullet$ we have $\Upsilon\vec{\zeta} = \vec{\zeta}$.

We now discuss the derivation of the resolvent formulas displayed in equation (2.10). Recall that $\vec{E}_0$ and $\vec{J}_0$ are constant vectors and that $\vec{E}_f \in \mathscr{H}_\times$ and $\vec{J}_f \in \mathscr{H}_\bullet$, so that $\Gamma\vec{E}_f = \vec{E}_f$ and $\Upsilon\vec{J}_f = \vec{J}_f$. Therefore, applying the operator $\nabla\Delta^{-1}$ to $\vec{\nabla}\cdot\vec{J} = 0$ in the first line of equation (2.3) and the operator $\nabla\times\Delta^{-1}$ to $\vec{\nabla}\times\vec{E} = 0$ in the second line, and noting that $\sigma = \sigma_2(1-\chi_1/s)$ and $\rho = \sigma_1^{-1}(1-\chi_2/s)$, for example, we obtain

$$\vec{E}_f = \frac{1}{s}\Gamma\chi_1\vec{E} = \frac{1}{t}\Gamma\chi_2\vec{E}, \qquad \vec{J}_f = \frac{1}{s}\Upsilon\chi_2\vec{J} = \frac{1}{t}\Upsilon\chi_1\vec{J}. \tag{2.11}$$

Equation (2.10) follows from (2.11) and the formulas $\vec{E} = \vec{E}_0 + \vec{E}_f$ and $\vec{J} = \vec{J}_0 + \vec{J}_f$.

It is more convenient to consider the functions $F(s) = 1 - m(h)$ and $E(s) = 1 - \tilde{m}(h)$, which are analytic off $[0,1]$ in the s-plane, and $G(t) = 1 - w(z)$ and $H(t) = 1 - \tilde{w}(z)$, which are analytic off $[0,1]$ in the t-plane [10, 65]. By equation (2.6) they satisfy

$$0 < |F(s)|, |E(s)| < 1, \quad 0 < |G(t)|, |H(t)| < \infty, \quad h \in \mathcal{U}_0. \tag{2.12}$$

We write $\vec{E}_0 = E_0\,\vec{e}_k$ and $\vec{J}_0 = J_0\,\vec{j}_k$, where $\vec{e}_k$ and $\vec{j}_k$ are standard basis vectors, for some $k = 1,\ldots,d$. Using equations (2.3), (2.5), (2.10), and the spectral theorem [101], we obtain the following integral representations of $F(s)$, $E(s)$, $G(t)$, and $H(t)$ [10, 12, 65]

$$\begin{aligned}
F(s) &= \langle \chi_1(s-\Gamma\chi_1)^{-1}\vec{e}_k\cdot\vec{e}_k\rangle = \int_{\lambda_0}^{\lambda_1}\frac{d\mu(\lambda)}{s-\lambda}\,, \\
E(s) &= \langle \chi_2(s-\Upsilon\chi_2)^{-1}\vec{j}_k\cdot\vec{j}_k\rangle = \int_{\tilde{\lambda}_0}^{\tilde{\lambda}_1}\frac{d\eta(\lambda)}{s-\lambda}\,, \\
G(t) &= \langle \chi_2(t-\Gamma\chi_2)^{-1}\vec{e}_k\cdot\vec{e}_k\rangle = \int_{\hat{\lambda}_0}^{\hat{\lambda}_1}\frac{d\alpha(\lambda)}{t-\lambda}\,, \\
H(t) &= \langle \chi_1(t-\Upsilon\chi_1)^{-1}\vec{j}_k\cdot\vec{j}_k\rangle = \int_{\check{\lambda}_0}^{\check{\lambda}_1}\frac{d\kappa(\lambda)}{t-\lambda}\,,
\end{aligned} \tag{2.13}$$

or in the compact notation of (2.9), $F(s) = \mathcal{A}_{0,1}(s;\mu)$, $E(s) = \mathcal{A}_{0,1}(s;\eta)$, $G(t) = \mathcal{A}_{0,1}(t;\alpha)$, and $H(t) = \mathcal{A}_{0,1}(t;\kappa)$. Equation (2.13) displays Stieltjes transforms of the bounded positive measures μ, η, α, and κ which are supported on $\Sigma_\mu, \Sigma_\eta, \Sigma_\alpha, \Sigma_\kappa \subseteq [0,1]$, respectively, and depend only on the geometry of the medium [12, 65]. The supremum and infimum of these sets are defined to be the upper and lower limits of integration displayed in equation (2.13).

The integro-differential operators $M_j = \chi_j\Gamma\chi_j$ and $K_j = \chi_j\Upsilon\chi_j$, $j = 1,2$, are compositions of projection operators on the associated Hilbert spaces $\mathscr{H}_\times$ and $\mathscr{H}_\bullet$, respectively, and are consequently positive definite and bounded by 1 in the underlying operator norm [104]. They are self-adjoint on $L^2(\Omega, P)$ [65]. Consequently, in the Hilbert space $L^2(\Omega, P)$ with weight χ_2 in the inner product, for example, $\Gamma\chi_2$ is a bounded self-adjoint operator [65]. Equation (2.13) is based upon spectral representations of resolvents involving these self-adjoint operators. The measures μ, η, α, and κ are spectral measures of the family of projections of these operators in the respective $\langle\vec{e}_k, \vec{e}_k\rangle$ or $\langle\vec{j}_k, \vec{j}_k\rangle$ state [65, 101].

A key feature of equations (2.4), (2.5), and (2.13) is that the parameter information in s and E_0 is *separated* from the geometry of the composite, which is encapsulated in the measures μ, η, α, and κ through their moments μ_n, η_n, α_n, and κ_n, $n \geq 0$, respectively, which depend on the correlation functions of the medium [65]. For example, $\alpha_0 = \eta_0 = p$ and $\mu_0 = \kappa_0 = 1-p$. A principal application of the analytic continuation method is to derive *forward bounds* on σ^* and ρ^*, given partial information on the microgeometry [11, 12, 65, 86]. One can also use the representations in (2.13) to obtain *inverse bounds*, allowing one to use data about the electromagnetic response of a sample to bound its structural parameters such as p [19, 31–34, 40, 64, 131].

We conclude this section by demonstrating that the energy constraints $\langle \vec{J}\cdot\vec{E}_f\rangle = \langle \vec{E}\cdot\vec{J}_f\rangle = 0$ lead to detailed decompositions of the system energy in terms of Herglotz functions involving μ, η, α, and κ. For example, the formulas $\langle \vec{J}\cdot\vec{E}_f\rangle = 0$, $\vec{E} = \vec{E}_0 + \vec{E}_f$, $\langle \vec{E}_f\rangle = 0$, and $\sigma = \sigma_2(1-\chi_1/s)$ imply that

$$0 = \langle \sigma\vec{E}\cdot\vec{E}_f\rangle = \langle \sigma_2(1-\chi_1/s)(\vec{E}_f\cdot\vec{E}_0 + E_f^2)\rangle = \sigma_2\left(\langle E_f^2\rangle - \frac{1}{s}\left(\langle \chi_1\vec{E}_f\cdot\vec{E}_0\rangle + \langle \chi_1 E_f^2\rangle\right)\right).$$

Denote by $R_s = (s-\Gamma\chi_1)^{-1}$ the resolvent of the operator $\Gamma\chi_1$. As $\Gamma\chi_1$ is self-adjoint in the $L^2(\Omega, P)$ inner product weighted by χ_1 [65], R_s is also self-adjoint in this inner product [118] for $s \in \mathbb{C}\backslash[0,1]$. Writing $\vec{E} = sR_s\vec{E}_0$ and $\vec{E}_0 = E_0\vec{e}_k$, the spectral theorem [101] then yields

$$\begin{aligned}\langle E_f^2\rangle &= \frac{E_0^2}{s}\left(\langle \chi_1(sR_s\vec{e}_k\cdot\vec{e}_k - 1)\rangle + \langle \chi_1\|(sR_s-1)\vec{e}_k\|^2\rangle\right) \\ &= \frac{E_0^2}{s}\left(\int_0^1\left[\frac{s}{s-\lambda}-1\right]d\mu(\lambda) + \int_0^1\left[\frac{s}{s-\lambda}-1\right]^2 d\mu(\lambda)\right) \\ &= \frac{E_0^2}{s}\int_0^1\frac{\lambda(s-\lambda)+\lambda^2}{(s-\lambda)^2}d\mu(\lambda) = E_0^2\int_0^1\frac{\lambda d\mu(\lambda)}{(s-\lambda)^2} = \mathcal{A}_{1,2}(s;\mu),\end{aligned} \tag{2.14}$$

where $\mathcal{A}_{i,j}(s;\mu)$ is defined in equation (2.9). By the symmetries of (2.13), we then have

$$\langle E_f^2\rangle/E_0^2 = \mathcal{A}_{1,2}(s;\mu) = \mathcal{A}_{1,2}(t;\alpha), \qquad \langle J_f^2\rangle/J_0^2 = \mathcal{A}_{1,2}(s;\eta) = \mathcal{A}_{1,2}(t;\kappa). \tag{2.15}$$

In Theorem 4 we show that equation (2.15) leads to Herglotz representations of all such energy components involving μ, η, α, and κ, We will focus on μ, as results associated with α, η and κ follow by symmetry.

Theorem 4 *Let $\mathcal{A}_{i,j}(s;\mu)$ be defined as in equation* (2.9)*. Then* (2.15) *implies that*

$$\begin{aligned}&\langle \chi_1 E_f^2\rangle/E_0^2 = \mathcal{A}_{2,2}(s;\mu), && \langle E_f^2\rangle/E_0^2 = \mathcal{A}_{1,2}(s;\mu), \\ &\langle \chi_1\vec{E}_f\cdot\vec{E}_0\rangle/E_0^2 = \mathcal{A}_{1,1}(s;\mu), && \langle \vec{E}_f\cdot\vec{E}_0\rangle/E_0^2 = 0, \\ &\langle \chi_1 E^2\rangle/E_0^2 = s^2\mathcal{A}_{0,2}(s;\mu), && \langle E^2\rangle/E_0^2 = 1+\mathcal{A}_{1,2}(s;\mu), \\ &\langle \chi_1\vec{E}\cdot\vec{E}_0\rangle/E_0^2 = s\mathcal{A}_{0,1}(s;\mu), && \langle \vec{E}\cdot\vec{E}_0\rangle/E_0^2 = 1, \\ &\langle \chi_1\vec{E}\cdot\vec{E}_f\rangle/E_0^2 = s^2\mathcal{A}_{0,2}(s;\mu) - s\mathcal{A}_{0,1}(s;\mu), && \langle \vec{E}\cdot\vec{E}_f\rangle/E_0^2 = \mathcal{A}_{1,2}(s;\mu),\end{aligned} \tag{2.16}$$

where the formulas involving χ_2 are given by the relation $\chi_2 = 1-\chi_1$ and (2.16).

Proof: Using equation (2.14) and the spectral theorem [101] we have

$$\frac{\langle \chi_1 E_f^2 \rangle}{E_0^2} = \langle \chi_1 (sR_s - 1)^2 \vec{e}_k \cdot \vec{e}_k \rangle = \int_0^1 \left(\frac{s}{s-\lambda} - 1 \right)^2 d\mu(\lambda) = \int_0^1 \frac{\lambda^2 d\mu(\lambda)}{(s-\lambda)^2} = \mathcal{A}_{2,2}(s;\mu),$$

$$\frac{\langle E_f^2 \rangle}{E_0^2} = \langle \| (sR_s - 1)\vec{e}_k \|^2 \rangle = \int_0^1 \frac{\lambda d\mu(\lambda)}{(s-\lambda)^2} = \mathcal{A}_{1,2}(s;\mu),$$

$$\frac{\langle \chi_1 \vec{E}_f \cdot \vec{E}_0 \rangle}{E_0^2} = \langle \chi_1 (sR_s - 1) \vec{e}_k \cdot \vec{e}_k \rangle = \int_0^1 \frac{\lambda d\mu(\lambda)}{s-\lambda} = \mathcal{A}_{1,1}(s;\mu),$$

$$\frac{\langle \vec{E}_f \cdot \vec{E}_0 \rangle}{E_0^2} = 0.$$

$$\frac{\langle \chi_1 E^2 \rangle}{E_0^2} = \langle \chi_1 \| sR_s \vec{e}_k \|^2 \rangle = \langle \chi_1 s^2 R_s^2 \vec{e}_k \cdot \vec{e}_k \rangle = s^2 \int_0^1 \frac{d\mu(\lambda)}{(s-\lambda)^2} = s^2 \mathcal{A}_{0,2}(s;\mu),$$

$$\frac{\langle E^2 \rangle}{E_0^2} = \frac{\langle (E_0^2 + 2\vec{E}_f \cdot \vec{E}_0 + E_f^2) \rangle}{E_0^2} = 1 + \int_0^1 \frac{\lambda d\mu(\lambda)}{(s-\lambda)^2} = 1 + \mathcal{A}_{1,2}(s;\mu),$$

$$\frac{\langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle}{E_0^2} = \langle \langle \chi_1 s R_s \vec{e}_k \cdot \vec{e}_k \rangle = s \int_0^1 \frac{d\mu(\lambda)}{s-\lambda} = s\mathcal{A}_{0,1}(s;\mu),$$

$$\frac{\langle \vec{E} \cdot \vec{E}_0 \rangle}{E_0^2} = 1.$$

$$\frac{\langle \chi_1 \vec{E} \cdot \vec{E}_f \rangle}{E_0^2} = \frac{\langle \chi_1 E^2 \rangle - \langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle}{E_0^2} = s^2 \mathcal{A}_{0,2}(s;\mu) - s\mathcal{A}_{0,1}(s;\mu),$$

$$\frac{\langle \vec{E} \cdot \vec{E}_f \rangle}{E_0^2} = \frac{\langle E^2 \rangle - \langle \vec{E} \cdot \vec{E}_0 \rangle}{E_0^2} = \int_0^1 \frac{\lambda d\mu(\lambda)}{(s-\lambda)^2} = \mathcal{A}_{1,2}(s;\mu),$$

These formulas hold for general two-component stationary random media in the lattice and continuum settings [60]. This concludes the proof of Theorem 4 □.

2.2 Stieltjes Function Representations of σ^* and ρ^*

In Section 2.1, we formulated the effective parameter problem for two-component conductive media and obtained integral representations of the effective complex conductivity σ^* and resistivity ρ^*. In this section we derive Stieltjes function representations of σ^* and ρ^*. These alternate representations will be used in Section 2.3 and Section 3.2 to provide spectral characterizations of critical behavior exhibited by σ^* and ρ^*.

In order to illuminate the many symmetries of this mathematical framework, we will henceforth focus on the complex variable $h = h_r + \mathrm{i}h_i$, where $h_r = \operatorname{Re} h$ and $h_i = \operatorname{Im} h$. Moreover, in the last two formulas of equation (2.13), we will make the change of variables $t(s) = 1 - s$ and $\lambda \mapsto 1 - \lambda$, so that

$$G(t(s)) = -\int_{1-\hat{\lambda}_1}^{1-\hat{\lambda}_0} \frac{[-d\alpha(1-\lambda)]}{s-\lambda}, \qquad H(t(s)) = -\int_{1-\check{\lambda}_1}^{1-\check{\lambda}_0} \frac{[-d\kappa(1-\lambda)]}{s-\lambda}.$$

The change of variables $s(h)=1/(1-h)$ and $\lambda(y)=y/(1+y) \iff y(\lambda)=\lambda/(1-\lambda)$ yield Stieltjes function representations [4] of the formulas in (2.13). For example,

$$F(s)=(1-h)\int_{S_0}^{S}\frac{(1+y)d\mu(\lambda(y))}{1+hy}\,,\quad G(t(s))=(h-1)\int_{\hat{S}_0}^{\hat{S}}\frac{(1+y)[-d\alpha(1-\lambda(y))]}{1+hy}\,, \tag{2.17}$$

where $S_0=\lambda_0/(1-\lambda_0)$, $S=\lambda_1/(1-\lambda_1)$, $\hat{S}_0=(1-\hat{\lambda}_1)/\hat{\lambda}_1$, $\hat{S}=(1-\hat{\lambda}_0)/\hat{\lambda}_0$, and the supports $\Sigma_\mu=[\lambda_0,\lambda_1]$ and $\Sigma_\alpha=[\hat{\lambda}_0,\hat{\lambda}_1]$ are defined in (2.13). Therefore, $\lim_{\lambda_0\longrightarrow 0}S_0=\lim_{\hat{\lambda}_1\longrightarrow 1}\hat{S}_0=0$ and $\lim_{\lambda_1\longrightarrow 1}S=\lim_{\hat{\lambda}_0\longrightarrow 0}\hat{S}=\infty$. Moreover, $d\mu(\lambda(y))$ is the measure $d\mu(\lambda)$ under the variable change $\lambda\mapsto\lambda(y)=y/(1+y)$ and $[-d\alpha(1-\lambda(y))]$ is the measure $d\alpha(\lambda)$ under the variable change $\lambda\mapsto 1-\lambda(y)$, where the negative sign accounts for the switch of integration limits in the second formula of (2.17). By equations (2.13) and (2.17), the Stieltjes function representations of $m(h)$ and $w(z(h))$ are given by

$$m(h)=1+(h-1)g(h),\quad g(h)=\int_0^\infty\frac{d\phi(y)}{1+hy}\,,\quad d\phi(y)=(1+y)d\mu(\lambda(y)), \tag{2.18}$$
$$w(z(h))=1-(h-1)\hat{g}(h),\quad \hat{g}(h)=\int_0^\infty\frac{d\hat{\phi}(y)}{1+hy}\,,\quad d\hat{\phi}(y)=(1+y)[-d\alpha(1-\lambda(y))],$$

and, by symmetry, analogous formulas for $\tilde{m}(h)$ and $\tilde{w}(z(h))$ involving Stieltjes functions $\tilde{g}(h)=\mathcal{S}_{0,1}(h;\tilde{\phi})$ and $\check{g}(h)=\mathcal{S}_{0,1}(h;\check{\phi})$, respectively. The Stieltjes functions $g(h),\tilde{g}(h),\hat{g}(h)$, and $\check{g}(h)$ are analytic for all $h\in\mathcal{U}_0$ [65]. As μ, η, α, and κ are positive measures on $[0,1]$, ϕ, $\tilde{\phi}$, $\hat{\phi}$, and $\check{\phi}$ are positive measures on $[0,\infty]$. Consequently, the following inequalities hold (see Lemma 1)

$$\frac{\partial^{2n}\zeta}{\partial h^{2n}}>0,\quad \frac{\partial^{2n+1}\zeta}{\partial h^{2n+1}}<0,\quad \left|\frac{\partial^n\zeta}{\partial h^n}\right|>0,\quad \zeta=g(h),\tilde{g}(h),\hat{g}(h),\check{g}(h),\quad h\in\mathcal{U}_0, \tag{2.19}$$

The first two inequalities in (2.19) hold for $h\in\mathcal{U}_0\cap\mathbb{R}$, $n\geq 0$, and the last inequality holds for $h\in\mathcal{U}_0\cap\mathbb{R}$ such that $h_i\neq 0$.

By equation (2.18), the moments ϕ_n of ϕ satisfy

$$\phi_n=\int_0^\infty y^n d\phi(y)=\int_0^\infty y^n(1+y)d\mu(\lambda)=\int_0^1\frac{\lambda^n d\mu(\lambda)}{(1-\lambda)^{n+1}}=\mathcal{A}_{n,n+1}(1;\mu)\,. \tag{2.20}$$

A partial fraction expansion of $\lambda^n/(1-\lambda)^{n+1}$ then shows that (see Lemma 1 below)

$$\frac{(-1)^n}{n!}\lim_{s\longrightarrow 1}\frac{\partial^n F(s)}{\partial s^n}=\int_0^1\frac{d\mu(\lambda)}{(1-\lambda)^{n+1}}=\sum_{j=0}^n\binom{n}{j}\phi_j\,. \tag{2.21}$$

Equation (2.21) demonstrates that ϕ_n depends on $\int_0^1 d\mu(\lambda)/(1-\lambda)^{n+1}$ *and* all the lower moments ϕ_j, $j=0,1,\ldots,n-1$, of ϕ. Equations (2.12) and (2.20) imply that ϕ_0 is bounded. In Lemma 2 below, we prove that the higher moments ϕ_n, $n\geq 1$, diverge as $\sup\{\Sigma_\mu\}\longrightarrow 1$.

We now show that Theorem 4 provides physical significance to the moments ϕ_j. Equations (2.13), (2.20), and (2.15) show that the first two moments, ϕ_0 and ϕ_1, of ϕ are identified with energy components:

$$\phi_0 = \lim_{s \longrightarrow 1} \frac{\langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle}{E_0^2}, \quad \phi_1 = \lim_{s \longrightarrow 1} \frac{\langle E_f^2 \rangle}{E_0^2}. \tag{2.22}$$

By equation (2.21), *all* of the higher moments ϕ_j, $j \geq 2$, depend on these energy components.

Similarly, the moments $\hat{\phi}_n$ of $\hat{\phi}$ satisfy (see Lemma 1)

$$\hat{\phi}_n = \int_0^1 \frac{(1-\lambda)^n d\alpha(\lambda)}{\lambda^{n+1}}, \qquad \frac{(-1)^{n+1}}{n!} \lim_{s \longrightarrow 1} \frac{\partial^n G(t(s))}{\partial^n t} = \int_0^1 \frac{d\alpha(\lambda)}{\lambda^{n+1}} = \sum_{j=0}^{n} \binom{n}{j} \hat{\phi}_j \,. \tag{2.23}$$

Equations (2.13), (2.15), and (2.23) also identify the first two moments, $\hat{\phi}_0$ and $\hat{\phi}_1$, of $\hat{\phi}$ with energy components. Equation (2.23) then implies that all of the higher moments $\hat{\phi}_j$, $j \geq 2$, depend on these energy components. We prove in Lemma 2 below that *all* the moments $\hat{\phi}_n$, $n \geq 0$, diverge as $\inf\{\Sigma_\alpha\} \longrightarrow 0$. By the symmetries in equations (2.13) and (2.18), equations (2.20) and (2.21) hold for $\tilde{\phi}$ with $E(s)$ and η in lieu of $F(s)$ and μ, respectively, and equation (2.23) holds for $\check{\phi}$ with $H(t(s))$ and κ in lieu of $G(t(s))$ and α, respectively.

We now give some key formulas that will be used extensively. Equations (2.4) and (2.5) yield the energy representations $\langle \vec{J} \cdot \vec{E} \rangle = \sigma_2 m(h) E_0^2 = \sigma_1 w(z(h)) E_0^2$ and $\langle \vec{E} \cdot \vec{J} \rangle = \tilde{m}(h) J_0^2/\sigma_1 = \tilde{w}(z(h)) J_0^2/\sigma_2$ involving σ^* and ρ^*, which imply that

$$m(h) = h w(z(h)) \iff 1 - F(s) = (1 - 1/s)(1 - G(t(s))), \quad h \in \mathcal{U}_0 \tag{2.24}$$

and an analogous formula linking $\tilde{m}(h)$ and $\tilde{w}(z(h))$. Equations (2.18) and (2.24) then yield

$$g(h) + h\hat{g}(h) = 1, \qquad \tilde{g}(h) + h\check{g}(h) = 1, \quad h \in \mathcal{U}_0. \tag{2.25}$$

For $h \in \mathcal{U}_0$, the functions $g(h)$, $\hat{g}(h)$, $\tilde{g}(h)$, and $\check{g}(h)$ are analytic [65] and have bounded h derivatives of all orders [104]. An inductive argument applied to equation (2.25) yields

$$\frac{\partial^n g}{\partial h^n} + n \frac{\partial^{n-1} \hat{g}}{\partial h^{n-1}} + h \frac{\partial^n \hat{g}}{\partial h^n} = 0, \qquad \frac{\partial^n \tilde{g}}{\partial h^n} + n \frac{\partial^{n-1} \check{g}}{\partial h^{n-1}} + h \frac{\partial^n \check{g}}{\partial h^n} = 0, \quad n \geq 1. \tag{2.26}$$

When $h \in \mathcal{U}_0$ such that $h_i \neq 0$, the complex representation of equation (2.26) is, for example,

$$\frac{\partial^n g_r}{\partial h^n} + n \frac{\partial^{n-1} \hat{g}_r}{\partial h^{n-1}} + h_r \frac{\partial^n \hat{g}_r}{\partial h^n} - h_i \frac{\partial^n \hat{g}_i}{\partial h^n} = 0, \quad \frac{\partial^n g_i}{\partial h^n} + n \frac{\partial^{n-1} \hat{g}_i}{\partial h^{n-1}} + h_r \frac{\partial^n \hat{g}_i}{\partial h^n} + h_i \frac{\partial^n \hat{g}_r}{\partial h^n} = 0,$$
$$\frac{\partial^n g_r}{\partial h^n} = \mathrm{Re} \frac{\partial^n g}{\partial h^n}, \quad \frac{\partial^n g_i}{\partial h^n} = \mathrm{Im} \frac{\partial^n g}{\partial h^n}, \qquad \frac{\partial^n \hat{g}_r}{\partial h^n} = \mathrm{Re} \frac{\partial^n \hat{g}}{\partial h^n}, \quad \frac{\partial^n \hat{g}_i}{\partial h^n} = \mathrm{Im} \frac{\partial^n \hat{g}}{\partial h^n}, \tag{2.27}$$

and analogous equations involving $\tilde{g}$ and $\check{g}$.

The integral representations of (2.26) and (2.27) follow from equation (2.28) of Lemma 1 below, involving the functions $\mathcal{S}_{i,j}$ defined in (2.9). In the remainder of this section, we focus on the measures ϕ and $\hat{\phi}$, as the analogous results involving $\tilde{\phi}$ and $\check{\phi}$ follow by symmetry.

Lemma 1 *For all $h \in \mathcal{U}_0$ and $i, j \in \mathbb{Z}$ satisfying $0 \le i \le j$, we have $|\mathcal{S}_{i,j}(h;\phi)| < \infty$, and for $0 \le i \le j-1$, $|\mathcal{S}_{i,j}(h;\hat{\phi})| < \infty$. Consequently ([55] Theorem 2.27), the Stieltjes functions $g(h)$ and $\hat{g}(h)$ may be repeatedly differentiated under the integral sign:*

$$\frac{\partial^n g(h)}{\partial h^n} = (-1)^n n! \int_0^\infty \frac{y^n d\phi(y)}{(1+hy)^{n+1}}\,, \qquad \frac{\partial^n \hat{g}(h)}{\partial h^n} = (-1)^n n! \int_0^\infty \frac{y^n d\hat{\phi}(y)}{(1+hy)^{n+1}}\,, \quad n \ge 0. \tag{2.28}$$

Before we prove Lemma 1, we note that equations (2.26) and (2.28) imply that

$$\int_0^\infty \frac{y^n d\phi(y)}{(1+hy)^{n+1}} = \int_0^\infty \frac{y^{n-1} d\hat{\phi}(y)}{(1+hy)^n} - h\int_0^\infty \frac{y^n d\hat{\phi}(y)}{(1+hy)^{n+1}}\,, \quad n \ge 1,\ h \in \mathcal{U}_0. \tag{2.29}$$

Moreover, equation (2.28) also yields the integral representations of (2.27) using

$$\frac{(-1)^n}{n!}\frac{\partial^n g(h)}{\partial h^n} = \int_0^\infty \frac{y^n d\phi(y)}{|1+hy|^{2(n+1)}}(1+\bar{h}y)^{n+1} = \sum_{j=0}^{n+1}\binom{n+1}{j}\bar{h}^j \int_0^\infty \frac{y^{n+j} d\phi(y)}{|1+hy|^{2(n+1)}}\,, \tag{2.30}$$

for example, where $\bar{h}$ denotes complex conjugation of the complex variable h.

Proof of Lemma 1 Let $\mathcal{S}_{i,j}(\xi;\nu)$ be defined as in equation (2.9). The supports of the measures ϕ and $\hat{\phi}$ are $\Sigma_\phi = [S_0, S\,]$ and $\Sigma_{\hat{\phi}} = [\hat{S}_0, \hat{S}\,]$, respectively, which are defined in terms of $\Sigma_\mu = [\lambda_0, \lambda_1]$ and $\Sigma_\alpha = [\hat{\lambda}_0, \hat{\lambda}_1]$, respectively, directly below equation (2.17). Recalling that $\lambda(y) = y/(1+y) \iff y(\lambda) = \lambda/(1-\lambda)$ and $s = 1/(1-h)$, equation (2.18) implies that

$$\mathcal{S}_{i,j}(h;\phi) = s^j \int_{\lambda_0}^{\lambda_1} \frac{\lambda^i (1-\lambda)^{j-i-1} d\mu(\lambda)}{(s-\lambda)^j}\,, \quad \mathcal{S}_{i,j}(h;\hat{\phi}) = s^j \int_{\hat{\lambda}_0}^{\hat{\lambda}_1} \frac{(1-\lambda)^i \lambda^{j-i-1} d\alpha(\lambda)}{(s-(1-\lambda))^j}\,. \tag{2.31}$$

We now show that $|\mathcal{S}_{i,j}(h;\phi)|$ and $|\mathcal{S}_{i,j}(h;\hat{\phi})|$ in (2.31) are uniformly bounded for all $h \in \mathcal{U}_\varepsilon$.

Set $0 < \varepsilon \ll 1$ and let $h \in \mathcal{U}_\varepsilon$, so that $|h| < 1$ and $|h - h_0| > \varepsilon$ for all $h_0 \in (-1, 0]$. As a complex variable, $s = 1/(1-h) = |s|^2(1 - h_r + \mathrm{i}h_i)$. Therefore, by the lower bound

$$|s|^2 = \frac{1}{|1-h|^2} = \frac{1}{1 - 2h_r + |h|^2} > \frac{1}{2(1-h_r)} > \frac{1}{2(1-\varepsilon)} > \frac{1}{2}\,, \tag{2.32}$$

when $h_i \neq 0$, we have $|s_i| = |s|^2|h_i| > \varepsilon/2 > 0$, and when $h_i = 0$, $s > 1/(1-\varepsilon) = 1 + \varepsilon/(1-\varepsilon) > 1 + \varepsilon$. Moreover, $t = 1 - s$ then implies that, when $h_i \neq 0$, $|t_i| > \varepsilon/2 > 0$, and when $h_i = 0$, $t < -\varepsilon < 0$. By equation (2.6) we also have $F(1) = \int_0^1 d\mu(\lambda)/(1-\lambda) \in [0,1]$. Thus, for $i, j \in \mathbb{Z}$ such that $0 \leq i \leq j$ and $0 \leq i \leq j-1$, equation (2.31) now implies that [104]

$$|\mathcal{S}_{i,j}(h;\phi)| \leq |s|^j \, \lambda_1^i \, (1-\lambda_0)^{j-i} \, F(1) \sup_{\lambda \in [\lambda_0, \lambda_1]} |s-\lambda|^{-j} < \infty \quad \text{and} \tag{2.33}$$
$$|\mathcal{S}_{i,j}(h;\hat{\phi})| \leq |s|^j \, (1-\hat{\lambda}_0)^i \, \hat{\lambda}_1^{j-i-1} \, \alpha_0 \sup_{\lambda \in [\hat{\lambda}_0, \hat{\lambda}_1]} |t-\lambda|^{-j} < \infty,$$

respectively, where α_0 is the mass of the measure α. We stress that these bounds, hence (2.28) hold even if $\lambda_0 = \hat{\lambda}_0 = 0$ and $\lambda_1 = \hat{\lambda}_1 = 1$ when $h \in \mathcal{U}_\varepsilon$. Because $g(h)$ and $\hat{g}(h)$ are analytic on $\mathcal{U}_0$ [65], they have bounded derivatives of all orders $n = 1, 2, 3, ...$ [104], and we may now let $\varepsilon \longrightarrow 0$ $\square$.

All of the equations given in this section display general formulas holding for two-component stationary random media in lattice and continuum settings [60]. In Section 3.2, we will investigate a class of composites for which the transport properties of σ^* exhibit critical behavior in the limit $|h| \longrightarrow 0$ ($|s| \longrightarrow 1$, $|t| \longrightarrow 0$). When $h = 0$, the bounds in (2.33) are violated when $\lambda_1 = 1$ and $\hat{\lambda}_0 = 0$, respectively. However, there is a class of composites for which there are *gaps* in the support of the measures μ and α about the spectral endpoints $\lambda = 0, 1$, such as matrix/particle composites [23], and the composite underlying effective medium theory (EMT) [87] (see, for example, Section 3.2.1). For such composite media, the bounds (2.33) and equation (2.28) are valid for $h \in \mathcal{U}_0 \cup \{0\}$. However, we will show in Section 2.3 that the limit of equation (2.29) as $h \longrightarrow 0$ is more subtle than simply setting $h = 0$.

While in general the spectra of μ and α extend all the way to $\lambda = 0, 1$, it has been argued that there are composites for which the spectrum close to $\lambda = 0, 1$ give exponentially small contributions to the transport properties of σ^* as $h \longrightarrow 0$ (Lifshitz phenomenon) [37, 75]. However, in this case, $|\partial^n g(h)/\partial h^n|$ and $|\partial^n \hat{g}(h)/\partial h^n|$ may diverge as $h \longrightarrow 0$ for some $n \geq 1$. We make this situation more precise by introducing a class $\mathcal{B}_{n_\nu}$ of composites in Definition 1 below.

Definition 1 *Define $\mathcal{B}_{n_\nu}$ to be the class of composites such that the functions $\mathcal{S}_{i,j}(h;\nu)$ in* (2.9) *satisfy $\lim_{|h| \longrightarrow 0} |\mathcal{S}_{n,n+1}(h;\nu)| < \infty$, where $\nu = \phi, \hat{\phi}$, for all $0 \leq n \leq n_\nu$.*

By equation (2.29), we have $n_\phi \geq n_{\hat{\phi}}$. The class $\mathcal{B}_{n_\nu}$, $\nu = \phi, \hat{\phi}$, will be used extensively in the proof of Theorem 5 below.

2.3 Spectral Characterization of Criticality in Transport

In this section we construct measures ϱ and $\tilde{\varrho}$ that are supported on $\{0,1\}$ which link the measures μ and α, and η and κ, respectively. The properties of ϱ and $\tilde{\varrho}$ imply that critical transitions in the transport properties of σ^* and ρ^* are due to the formation of delta function components in the underlying spectral measures at $\lambda = 0, 1$. In Section 3.2, this identifies these transport transitions with the collapse of spectral gaps in these measures and leads to a precise spectral characterization of critical transport behavior in binary composite media.

The Stieltjes transform of the spectral measures μ, α, η, and κ completely determines the effective transport properties of the medium. Conversely, given the Stieltjes transform of a measure, the Stieltjes–Perron Inversion Theorem [71] recovers the underlying measure,

$$\mu(\upsilon) = -\frac{1}{\pi}\lim_{\epsilon\downarrow 0}\operatorname{Im} F(\upsilon + \mathrm{i}\epsilon)\ , \quad \upsilon \in \Sigma_\mu, \tag{2.34}$$

for example. To evoke this theorem directly, in equation (2.13) we define the measures $d\tilde{\alpha}(\lambda) = [-d\alpha(1-\lambda)]$ and $d\tilde{\kappa}(\lambda) = [-d\kappa(1-\lambda)]$, and write $G(t(s)) = -\int_0^1 d\tilde{\alpha}(\lambda)/(s-\lambda)$ and $H(t(s)) = -\int_0^1 d\tilde{\kappa}(\lambda)/(s-\lambda)$. Setting $s = \upsilon + \mathrm{i}\epsilon$, equations (2.24) and (2.34) imply that

$$\upsilon\mu(\upsilon) = (1-\upsilon)[-\alpha(1-\upsilon)] - \upsilon\varrho(\upsilon), \qquad \varrho(\upsilon) = \lim_{\epsilon\downarrow 0}\frac{-\epsilon/\pi}{\upsilon^2+\epsilon^2}\int_0^1 \frac{(\upsilon+\lambda-1)\,d\alpha(\lambda)}{(\upsilon+\lambda-1)^2+\epsilon^2}\ , \tag{2.35}$$

and an analogous formula involving a measure $\tilde{\varrho}$ which links η and κ. We now demonstrate that equations (2.24), (2.25), and (2.35) explicitly determine the measures ϱ and $\tilde{\varrho}$.

The integral representations of equation (2.25) follow from equation (2.18), and are given by

$$\int_0^\infty \frac{d\phi(y)}{1+hy} + h\int_0^\infty \frac{d\hat{\phi}(y)}{1+hy} = 1, \qquad \int_0^\infty \frac{d\tilde{\phi}(y)}{1+hy} + h\int_0^\infty \frac{d\check{\phi}(y)}{1+hy} = 1. \tag{2.36}$$

Due to the underlying symmetries of this framework, without loss of generality, we henceforth focus on $F(s(h);\mu)$, $G(t(h);\alpha)$, $g(h;\phi)$, and $\hat{g}(h;\hat{\phi})$. We wish to re-express the first formula in equation (2.36) in a more suggestive form by adding and subtracting the quantity $h\int_0^\infty y\,d\phi(y)/(1+hy)$. This is permissible if the modulus of this quantity is finite for all $h \in \mathcal{U}_0$ [55, 104]. The affirmation of this fact is given by Lemma 1 and we may therefore add and subtract it in equation (2.36), yielding

$$h\int_0^\infty \frac{d\Phi_0(y)}{1+hy} \equiv 1 - \phi_0 = m(0), \quad d\Phi_0(y) = d\hat{\phi}(y) - y\,d\phi(y), \quad h \in \mathcal{U}_0, \tag{2.37}$$

as $1-\phi_0 = 1-F(s)|_{s=1} = m(h)|_{h=0}$ (see equation (2.20)). We stress that $\sigma_1 \neq \sigma_2$ when $h = 0$ so that in (2.6) $0 \leq m(0) < 1$. Equation (2.37) provides another representation for the quantity $m(0)$ and shows that the transform $h\int_0^\infty d\Phi_0(y)/(1+hy)$ of Φ_0, a signed measure [104], is independent of h for all $h \in \mathcal{U}_0$. Equation (2.18) and the identity $y = \lambda/(1-\lambda) \iff \lambda = y/(1+y)$ relates this representation of $m(0)$ to the measure ϱ defined in equation (2.35):

$$d\Phi_0(y) = \frac{1}{(1-\lambda)^2}((1-\lambda)\,[-d\alpha(1-\lambda)] - \lambda\, d\mu(\lambda)) = \frac{\lambda\, d\varrho(\lambda)}{(1-\lambda)^2} = y(1+y)\, d\varrho\,(\lambda(y))\,.$$

We may now express equation (2.37) in terms of $\varrho(d\lambda)$ as follows:

$$m(0) = h\int_0^\infty \frac{d\Phi_0(y)}{1+hy} = h\int_0^\infty \frac{y(1+y)d\varrho(\lambda(y))}{1+hy} = \int_0^1 \frac{\lambda\, d\varrho(\lambda)}{(1-\lambda)^2/h + \lambda(1-\lambda)}\,. \tag{2.38}$$

Remark 1 *Define the transform $\mathcal{D}(h;\varrho)$ of the measure ϱ by*

$$\mathcal{D}(h;\varrho) = \int_0^1 \frac{\lambda\, d\varrho(\lambda)}{(1-\lambda)^2/h + \lambda(1-\lambda)}\,. \tag{2.39}$$

*Equations (2.6) and (2.38) show that $\mathcal{D}(h;\varrho)$ has the following properties for all $h \in \mathcal{U}_0$: **(1)** $\mathcal{D}(h;\varrho)$ is independent of h, **(2)** $0 \leq |\mathcal{D}(h;\varrho)| < 1$, and **(3)** $\mathcal{D}(h;\varrho) = m(0) \not\equiv 0$.*

Lemma 2 *Let the quantities $m(0) = m(h)|_{h=0} = 1-F(s)|_{s=1}$ and $w(0) = w(z)|_{z=0} = 1-G(t)|_{t=1}$ be defined as in equation (2.13), which satisfy $0 \leq m(0), w(0) < 1$. If $\mathcal{D}(h;\varrho)$, defined in equation (2.39), satisfies the properties of Remark 1 for all $h \in \mathcal{U}_0$, then*

$$\varrho(d\lambda) = -w(0)\delta_0(d\lambda) + m(0)(1-\lambda)\delta_1(d\lambda), \tag{2.40}$$

where $\delta_{\lambda_0}(d\lambda)$ is the Dirac measure concentrated at λ_0.

Proof: Let $\mathcal{D}(h;\varrho)$, defined in equation (2.39), satisfy properties (**1**)–(**3**) of Remark 1. The measure ϱ is independent of h [65]. If the support Σ_ϱ of the measure ϱ is over continuous spectrum [101] then $\mathcal{D}(h;\varrho)$ depends on h, contradicting property (**1**). Therefore the measure ϱ is defined over pure point spectrum [101]. The most general pure point set Σ_ϱ which satisfies properties (**1**)–(**3**) is given by $\Sigma_\varrho = \{0,1\}$. This implies that the measure ϱ is of the form

$$\varrho(d\lambda) = W_0(\lambda)\delta_0(d\lambda) + W_1(\lambda)\delta_1(d\lambda),$$

where the $W_j(\lambda)$, $j = 0,1$, are bounded functions of $\lambda \in [0,1]$ which are to be determined. In view of the numerator of the integrand in equation (2.39), we may assume that the function $W_0(\lambda) \equiv W_0(0) = W_0 \not\equiv 0$ is independent of λ. In order for properties (**2**) and (**3**) to be satisfied we must have $W_1(\lambda) \sim (1-\lambda)^1$ as $\lambda \longrightarrow 1$ (any other power of $1-\lambda$ would contradict one of these two properties). Therefore without loss of generality, we may set $W_1(\lambda) = \mathrm{w}_1\,(1-\lambda)$, where w_1 is independent of λ. Property (**3**) now yields $\mathrm{w}_1 = m(0)$.

We have shown that $\varrho(d\lambda) = W_0\,\delta_0(d\lambda) + m(0)(1-\lambda)\delta_1(d\lambda)$, $W_0 \not\equiv 0$. By plugging this formula into equation (2.35) ($\lambda d\mu(\lambda) = (1-\lambda)[-d\alpha(1-\lambda)] - \lambda d\varrho(\lambda)$), we are able determine W_0. Indeed using equation (2.24) ($F(s) - (1-1/s)G(t(s)) = 1/s$), the definition of $F(s)$ in equation (2.13), and $(1-\lambda)/(\lambda(s-\lambda)) = -(1-1/s)/(s-\lambda) + 1/(s\lambda)$, we find that

$$\begin{aligned} F(s) &= -\left(1-\frac{1}{s}\right)\int_0^1 \frac{[-d\alpha(1-\lambda)]}{s-\lambda} + \frac{1}{s}\int_0^1 \frac{[-d\alpha(1-\lambda)]}{\lambda} - \int_0^1 \frac{d\varrho(\lambda)}{s-\lambda} \\ &= \left(1-\frac{1}{s}\right)G(t(s)) + \frac{1}{s}\int_0^1 \frac{d\alpha(\lambda)}{1-\lambda} - \frac{W_0}{s} - m(0)\lim_{\lambda\longrightarrow 1}\frac{1-\lambda}{s-\lambda}, \quad \forall\, h \in \mathcal{U}_0 \end{aligned} \tag{2.41}$$

which implies that $-W_0 = 1 - \int_0^1 d\alpha(\lambda)/(1-\lambda) = w(0)$ □.

Corollary 1 *If we instead focus on the contrast variables z and t in lieu of h and s, respectively, equations* (2.35) *and* (2.40) *become*

$$\upsilon\alpha(\upsilon) = (1-\upsilon)[-\mu(1-\upsilon)] - \upsilon\varrho(\upsilon), \quad \varrho(d\lambda) = -m(0)\delta_0(d\lambda) + w(0)(1-\lambda)\delta_1(d\lambda), \tag{2.42}$$

It is worth mentioning that equation (2.29) can be written in terms of the signed measure $d\Phi_{n-1}(y) = y^{n-1}d\Phi_0(y)$: $\int_0^\infty d\Phi_{n-1}(y)/(1+hy)^{n+1} \equiv 0$, for all $n \geq 1$. Furthermore in equation (2.27) for $n = 1$, equation (2.30) implies that $\int_0^\infty d\Phi_1(y)/|1+hy|^4 \equiv 0$. By Lemma 1, these integral involving $\Phi_{n-1}(dy)$ are defined for all $h \in \mathcal{U}_0$. These formulas are consistent with equation (2.40) of Lemma 2.

Lemma 2 and Corollary 1 are the key results of this section. They provide a rigorous justification, and a generalization of an analogous result found in [39] by heuristic means. They demonstrate that $\lambda = 1$ is a removable *simple* singularity under μ, α, η, and κ, and illustrate how the relations in (2.12), $0 < |F(s)|, |E(s)\ | < 1$, can hold even when $s = 1$ ($h = 0$) and the spectra extend all the way to $\lambda = 1$. For percolation models, Lemma 2 and Corollary 1 also demonstrate that delta components form in the underlying spectral measures at the spectral endpoints precisely at the percolation threshold p_c and $1-p_c$. This is analogous to the Lee–Yang–Ruelle characterization of the Ising model phase transition, and identifies these transport transitions with the collapse of spectral gaps in these measures. In Section 3.2, we discuss in more detail how these general features relate to percolation models of binary composite media.

CHAPTER 3

CRITICAL BEHAVIOR OF TRANSPORT IN BINARY COMPOSITE MEDIA

In this chapter we construct a mathematical framework which unifies the critical theory of transport in two-phase random media. By adapting techniques developed by G. A. Baker for the Ising model [4], we provide a detailed description of percolation-driven critical transitions in transport exhibited by such media. The most natural formulation is in terms of the conduction problem in the continuum $\mathbb{R}^d$, which includes the lattice $\mathbb{Z}^d$ as a special case [59, 65]. Although, symmetries in Maxwell's equations [87] immediately extend our results to the effective parameter problem of electrical permittivity and the critical behavior of two-phase random dielectric media.

An original motivation for this work was to gain a better understanding of critical transitions in the transport properties of sea ice. In particular, fluid flow through sea ice mediates a broad range of processes that are important to studying its role in the climate system, and the impact of climate change on polar ecosystems [61]. In fact, the brine microstructure of sea ice displays a percolation threshold at a critical brine volume fraction ϕ of about 5% in columnar sea ice [62, 63, 98]. This leads to critical behavior of fluid flow, where sea ice is effectively impermeable to fluid transport for ϕ below 5%, and is increasingly permeable for ϕ above 5%, which is known as the *rule of fives* [62]. Percolation theory can then be used to capture the behavior of the fluid permeability of sea ice [63]. There has also been evidence [68, 95] that this critical behavior in the microstructure also induces similar behavior in the effective electromagnetic properties of sea ice, such as its effective complex permittivity ϵ^*. In [68] and [95], for example, microstructural properties of the brine phase were recovered from measurements of the complex permittivity of sea ice. The current chapter helps lay the groundwork for the analysis of sea ice permittivity data collected in the polar regions, and how it can be used to monitor changes in the microstructure, the fluid transport properties, and the geophysical and biological processes that are controlled by fluid flow.

3.1 Background and Summary of the Results

The partition function Z of the Ising model is a polynomial in the activity variable [4, 81, 105, 107]. In 1952, Lee and Yang [81] showed that the roots of Z lie on the unit circle, which is known as the Lee–Yang Theorem [81, 105]. They also demonstrated that the distribution of the roots determines the associated equation of state [130], and that the properties of the system, in relation to phase transitions, are governed by the behavior of these roots near the positive real axis.

In 1968 Baker [3] used the Lee–Yang Theorem to represent the Gibbs free energy per spin $f = -(N\beta)^{-1} \ln Z$ as a logarithmic potential [108], where N is the number of spins, $\beta = (kT)^{-1}$, k is Boltzmann's constant, and T is the absolute temperature. He used this special analytic structure to prove that the magnetization per spin $M(T, H) = -\partial f/\partial H$ [103] may be represented in terms of a Stieltjes function G in the variable $\tau = \tanh \beta m H$,

$$\frac{M}{m} = \tau(1 + (1 - \tau^2)G(\tau^2)), \quad G(\tau^2) = \int_0^\infty \frac{d\psi(y)}{1 + \tau^2 y}\,, \tag{3.1}$$

where H is the applied magnetic field strength, m is the (constant) magnetic dipole moment of each spin [66], and ψ is a nonnegative definite measure [3, 4]. Equation (3.1) should be compared to equation (2.18) regarding the two-phase conductive media. The integral representation in (3.1) immediately leads to the inequalities

$$G \geq 0, \qquad \frac{\partial G}{\partial u} \leq 0, \qquad \frac{\partial^2 G}{\partial u^2} \geq 0, \tag{3.2}$$

where $u = \tau^2$, which are analogs of equation (2.19) for two-phase conductive media [59]. The last formula in equation (3.2) is the GHS inequality, which is an important tool in the study of the Ising model [4, 59].

In 1970, Ruelle [106] extended the Lee–Yang Theorem and proved that there exists a *gap* $\theta_0(T) > 0$ in the roots of Z about the positive real axis for high temperatures. Moreover, he proved that the gap collapses, $\theta_0(T) \longrightarrow 0$, as T decreases to a critical temperature $T_c > 0$. Consequently, the temperature-driven phase transition (spontaneous magnetization) is unique, and is characterized by the pinching of the real axis by the roots of Z [105].

Baker [4, 5] then exploited the Lee–Yang–Ruelle Theorem to provide a detailed description of the critical behavior of the parameters characterizing the phase transition exhibited by the Ising model [36]. He defined a critical exponent Δ for the gap in the distribution of the Lee–Yang–Ruelle zeros, $\theta_0(T) \sim (T - T_c)^\Delta$, as $T \longrightarrow T_c^+$, and proved that the measure ψ is supported on the compact interval $[0, S(T)]$ for $T > T_c$, with $S(T) \sim (T - T_c)^{-2\Delta}$ as

$T \longrightarrow T_c^+$. He demonstrated that the moments $\psi_n = \int_0^\infty y^n \, d\psi(y)$ of ψ diverge as $T \longrightarrow T_c^+$ according to the power law $\psi_n \sim (T - T_c)^{-\gamma_n}$, $n \geq 0$, by proving that the sequence γ_n satisfies Baker's inequalities $\gamma_{n+1} - 2\gamma_n + \gamma_{n-1} \geq 0$. They imply that this sequence increases at least linearly with n. He later proved that this sequence is actually linear in n, $\gamma_n = \gamma + 2\Delta n$, with constant gap $\gamma_i - \gamma_{i-1} = 2\Delta$ [4]. The critical exponent γ is defined via the magnetic susceptibility per spin $\chi = \partial M/\partial H = -\partial^2 f/\partial H^2 \sim (T - T_c)^{-\gamma}$, as $T \longrightarrow T_c^+$.

The phase transition may be concisely described with two other critical exponents. When $H = 0$, $M(T, 0) \sim (T - T_c)^\beta$, as $T \longrightarrow T_c^-$, where the critical exponent β is not to be confused with $(kT)^{-1}$, and along the critical isotherm $T = T_c$, $M(T_c, H) \sim H^{1/\delta}$, as $H \longrightarrow 0$ [4, 36]. Using the integral representation in (3.1), Baker obtained (two-parameter) scaling relations for these critical exponents [4]

$$\beta = \Delta - \gamma, \qquad \delta = \Delta/(\Delta - \gamma), \qquad \gamma_n = \gamma + 2\Delta n. \tag{3.3}$$

The critical exponent γ, for example, is defined in terms of the following limit, and γ exists when this limit exists [4],

$$\gamma = \limsup_{T \longrightarrow T_c^+,\ H=0} \left(\frac{-\ln \chi(T, H)}{\ln(T - T_c)} \right). \tag{3.4}$$

In 1997 Golden [60] demonstrated that Baker's critical theory may be adapted to provide a precise description of percolation-driven critical transitions in transport, exhibited by two-phase random media in the static regime. This result puts these two classes of seemingly unrelated problems on an equal mathematical footing. He did so by considering percolation models of classical conductive two-phase composite media, where the connectedness of the system is determined, for example, by the volume fraction p of inclusions with conductance σ_2 in an otherwise homogeneous medium of conductivity σ_1, with $h = \sigma_1/\sigma_2 \in [0, 1]$. He demonstrated that the function $m(p, h) = \sigma^*(p, h)/\sigma_2$ plays the role of the magnetization $M(T, H)$, where σ^* is the effective conductivity of the medium [10, 65, 86]. Moreover, he showed that the volume fraction p mimics the temperature T while the contrast ratio h is analogous to the applied magnetic field strength H. More specifically, critical behavior of transport arises when $h = 0$ ($\sigma_1 = 0,\ 0 < \sigma_2 < \infty$), as $p \longrightarrow p_c^+$ [60], and critical behavior of the magnetization in the Ising model arises when $H = 0$, as $T \longrightarrow T_c^+$ [36]. Using these mathematical parallels, it was shown that the critical exponents of transport satisfy an analogue of Baker's scaling relations (3.3).

Here, using a novel unified approach, we reproduce Golden's static results ($h \in \mathbb{R}$) and obtain the analogous static results associated with a conductive-superconductive medium in terms of $w(p, z) = \sigma^*(p, z)/\sigma_1$, where $z = 1/h$. Using Stieltjes function integral representations of $m(p, h; \mu)$ and $w(p, z; \alpha)$, where μ and α are each spectral measures of a random self-adjoint operator, we determine the (two-parameter) critical exponent scaling relations of each system. We then extend these results to the frequency dependent quasi-static regime ($h \in \mathbb{C}$). We also link these two sets of critical exponents and, assuming a symmetry in the properties of μ and α, the resultant scaling relations linking the two sets of critical exponents are in agreement with the seminal paper by A. L. Efros and B. I. Shklovskii [52]. We remark that there are similar critical exponents involving ϵ^* for two-phase dielectric media [13, 37], and there are direct analogs of our results regarding such media.

In arbitrary finite lattice systems we also explicitly show that there are gaps in the supports of the measures $\alpha(d\lambda)$ and $\mu(d\lambda)$ about the spectral endpoints $\lambda = 0, 1$ for $p \ll 1$ and $1 - p \ll 1$, respectively. Recall that in Section 2.3 we demonstrated, for infinite lattice or continuum composite systems, that critical transitions in transport are due to the formation of delta components in μ and α located at $\lambda = 0, 1$. We did so by constructing a measure ϱ which is supported on the set $\{0, 1\}$ that links μ and α. This general result demonstrates that, for percolation models, the onset of criticality (the formation of these delta components) occurs *precisely* at the percolation threshold p_c and at $1 - p_c$.

3.2 Scaling Laws for Critical Exponents of Transport in Lattice and Continuum Percolation Models

We now formulate the problem of percolation-driven critical transitions in transport exhibited by two-component conductive media. In modeling transport in such materials, one often considers a two-component random medium with component conductivities σ_1 and σ_2, in the volume fractions $1 - p$ and p, respectively. The medium may be continuous, like the random checkerboard [14, 112] and Swiss cheese models [13, 69, 116], or discrete, like the random resistor network (RRN) [13, 37, 116]. In the simplest case of the 2-d square RRN [116, 122], the average cluster size of the σ_2 inclusions grows as p increases, and there is a critical volume fraction p_c, $0 < p_c < 1$, called the *percolation threshold*, where an infinite cluster of σ_2 bonds first appears. In the limit $h = \sigma_1/\sigma_2 \longrightarrow 0$, the system exhibits two types of critical behavior. First, as $h \longrightarrow 0$ ($\sigma_1 \longrightarrow 0$ and $0 < |\sigma_2| < \infty$), the effective complex conductivity $\sigma^*(p, h) = \sigma_2\, m(p, h)$ and the effective complex resistivity $\rho^*(p, z(h)) = \tilde{w}(p, z(h))/\sigma_2$ undergo a conductor-insulator critical transition [13]:

$$|\sigma^*(p,0)| = 0, \quad \text{for} \quad p < p_c, \quad \text{and} \quad 0 = |\sigma_1| < |\sigma^*(p,0)| < |\sigma_2| < \infty, \quad \text{for} \quad p > p_c, \tag{3.5}$$
$$\lim_{p \longrightarrow p_c^+} |\rho^*(p,z(0))| = \infty, \quad \text{and} \quad 0 < |\sigma_2|^{-1} < |\rho^*(p,z(0))| < |\sigma_1|^{-1} = \infty, \quad \text{for} \quad p > p_c.$$

Second, as $h \longrightarrow 0$ ($|\sigma_2| \longrightarrow \infty$ and $0 < |\sigma_1| < \infty$), the effective complex conductivity $\sigma^*(p,z(h)) = \sigma_1 w(p,z(h))$ and the effective complex resistivity $\rho^*(p,h) = \tilde{m}(p,h)/\sigma_1$ undergo a conductor-superconductor critical transition [13]:

$$0 < |\sigma_1| < |\sigma^*(p,z(0))| < |\sigma_2| = \infty, \quad \text{for} \quad p < p_c, \quad \text{and} \quad \lim_{p \longrightarrow p_c^-} |\sigma^*(p,z(0))| = \infty. \tag{3.6}$$
$$0 = |\sigma_2|^{-1} < |\rho^*(p,0)| < |\sigma_1|^{-1} < \infty, \quad \text{for} \quad p < p_c, \quad \text{and} \quad |\rho^*(p,0)| = 0, \quad \text{for} \quad p > p_c.$$

We will focus on the conductor-insulator critical transition of the effective complex conductivity $\sigma^*(p,h) = \sigma_2\, m(p,h)$ and the conductor-superconductor critical transition of the effective complex conductivity $\sigma^*(p,z(h)) = \sigma_1 w(p,z(h))$. It is clear from equations (2.18), (3.5), and (3.6) that our results immediately generalize to $\rho^*(p,h) = \tilde{m}(p,h)/\sigma_1$ and $\rho^*(p,z(h)) = \tilde{w}(p,z(h))/\sigma_2$, respectively, with $p \mapsto 1-p$.

This critical behavior in transport is made more precise through the definition of critical exponents. Recall that the existence of a critical exponent is determined by the existence of a limit like that given in (3.4). In the static limit, $h \in \mathcal{U}_0 \cap \mathbb{R}$, as $h \longrightarrow 0$ the effective conductivity $\sigma^*(p,h) = \sigma_2\, m(p,h)$ exhibits critical behavior near the percolation threshold $\sigma^*(p,0) \sim (p-p_c)^t$, as $p \longrightarrow p_c^+$. Here, the critical exponent t, not to be confused with the contrast parameter, is believed to be *universal* for lattices, depending only on dimension [60]. At $p = p_c$, $\sigma^*(p_c,h) \sim h^{1/\delta}$ as $h \longrightarrow 0$. We assume the existence of the critical exponents t and δ, as well as γ, defined via a conductive susceptibility $\chi(p,0) = \partial m(p,0)/\partial h \sim (p-p_c)^{-\gamma}$, as $p \longrightarrow p_c^+$. For $p > p_c$, we assume that there is a gap $\theta_\mu \sim (p-p_c)^\Delta$ in the support of μ around $h = 0$ or $s = 1$ which collapses as $p \longrightarrow p_c^+$, or that any spectrum in this region does not affect power law behavior [60]. Consequently, for $p > p_c$ we think of the support of ϕ as being contained in the interval $[0, S(p)]$, with $S(p) \sim (p-p_c)^{-\Delta}$ as $p \longrightarrow p_c^+$. We demonstrated in (2.20) that the moments ϕ_j of ϕ become singular as $\theta_\mu \longrightarrow 0$. We therefore assume the existence of critical exponents γ_n such that $\phi_n(p) \sim (p-p_c)^{-\gamma_n}$ as $p \longrightarrow p_c^+$, $n \geq 0$. When $h \in \mathcal{U}_0$ such that $h_i \neq 0$, we also assume the existence of critical exponents t_r, δ_r, t_i and δ_i corresponding to $m_r(p,h) = \operatorname{Re} m(p,h)$ and $m_i(p,h) = \operatorname{Im} m(p,h)$. In summary:

$$m(p,0) \sim (p-p_c)^t, \quad m_r(p,0) \sim (p-p_c)^{t_r}, \quad m_i(p,0) \sim (p-p_c)^{t_i}, \quad \text{as} \;\; p \longrightarrow p_c^+ \tag{3.7}$$
$$m(p_c,h) \sim h^{1/\delta}, \quad m_r(p_c,h) \sim |h|^{1/\delta_r}, \quad m_i(p_c,h) \sim |h|^{1/\delta_i}, \quad \text{as} \;\; |h| \longrightarrow 0,$$
$$\chi(p,0) \sim (p-p_c)^{-\gamma}, \quad \phi_n \sim (p-p_c)^{-\gamma_n}, \quad S(p) \sim (p-p_c)^{-\Delta}, \quad \text{as} \;\; p \longrightarrow p_c^+.$$

In a similar way we define critical exponents for the conductor-superconductor system:

$$w(p) \sim (p_c-p)^{-s}, \quad w_r(p) \sim (p_c-p)^{-s_r}, \quad w_i(p) \sim (p_c-p)^{-s_i}, \quad \text{as } p \longrightarrow p_c^-$$
$$w(p_c,z(h)) \sim h^{-1/\hat{\delta}}, \quad w_r(p_c,z(h)) \sim |h|^{-1/\hat{\delta}_r}, \quad w_i(p_c,z(h)) \sim |h|^{-1/\hat{\delta}_i}, \quad \text{as } |h| \longrightarrow 0,$$
$$\hat{\chi}(p,z(0)) \sim (p_c-p)^{-\hat{\gamma}'}, \quad \hat{\phi}_n \sim (p_c-p)^{-\hat{\gamma}'_n}, \quad \hat{S}(p) \sim (p_c-p)^{-\hat{\Delta}'}, \quad \text{as } p \longrightarrow p_c^-, \tag{3.8}$$

where we have defined $w(p) = w(p,z(0))$ and s is the superconductor critical exponent, not to be confused with the contrast parameter. We also assume the existence of critical exponents γ', γ'_n, and Δ', associated with $m(p,h;\phi)$ for the left hand limit $p \longrightarrow p_c^-$, and critical exponents $\hat{\gamma}$, $\hat{\gamma}_n$, and $\hat{\Delta}$, associated with $w(p,z(h);\hat{\phi})$ for the right hand limit $p \longrightarrow p_c^+$. The critical exponents γ, δ, Δ, and γ_n for transport are different from those defined in Section 3.1 for the Ising model in (3.3).

The key result of this section is the two-parameter scaling relations between the critical exponents of the conductor-insulator system, defined in equation (3.7), and that of the conductor-superconductor system, defined in equation (3.8). Moreover, Lemma 2 shows that measures μ and α, hence ϕ and $\hat{\phi}$ are related, and we therefore anticipate that these two sets of critical exponents are also related. This is indeed the case and, assuming a symmetry in the properties of μ and α, the resultant relationship between the critical exponents t and s is in agreement with the seminal paper by A. L. Efros and B. I. Shklovskii [52].

These results are summarized in Theorem 5 below. In this theorem, we assume that the percolation model under consideration is of class $\mathcal{B}_{n_\phi}$ and $\mathcal{B}_{n_{\hat{\phi}}}$ for $p > p_c$ and $p < p_c$, respectively (see Definition 1). By equations (2.29), (3.5), and (3.6), the 2-d square RRN is of class $\mathcal{B}_{n_{\hat{\phi}}}$ and $\mathcal{B}_{n_\phi}$ for $p > p_c$ and $p < p_c$ with $0 \le n_{\hat{\phi}} \le n_\phi$. We assume that this holds for the percolation model under consideration, and we further assume that $1 \le n_{\hat{\phi}} \le n_\phi$ so that $\chi(p,0) = \partial m(p,0)/\partial h$ and $\hat{\chi}(p,z(0)) = \partial w(p,z(0))/\partial h$ in (3.7) and (3.8) exist.

Theorem 5 *Consider a percolation model of a binary conductive medium of class* $\mathcal{B}_{n_\phi}$ *and* $\mathcal{B}_{n_{\hat{\phi}}}$ *for* $p > p_c$ *and* $p < p_c$*, respectively, and* $1 \le n_{\hat{\phi}} \le n_\phi$*. Let the critical exponents associated with the model:* t, t_r, t_i, δ, δ_r, δ_i, γ, γ_n, Δ, γ', γ'_n, *and* Δ', *and* s, s_r, s_i, $\hat{\delta}$, $\hat{\delta}_r$,

$\hat{\delta}_i$, $\hat{\gamma}'$, $\hat{\gamma}'_n$, $\hat{\Delta}'$, $\hat{\gamma}$, $\hat{\gamma}_n$, and $\hat{\Delta}$, be defined as in equations (3.7) *and* (3.8)*, respectively, and in the paragraph following equation* (3.8)*. Then the following scaling relations hold:*

1) $\gamma_1 = \gamma,\ \gamma'_1 = \gamma',\ \hat{\gamma}_1 = \hat{\gamma},$ *and* $\hat{\gamma}'_1 = \hat{\gamma}'$.

2) $\gamma'_0 = 0,\ \gamma_0 < 0,\ \gamma'_n > 0$ *and* $\gamma_n > 0,\ n \geq 1$. **3)** $\hat{\gamma}'_n > 0$ *for* $n \geq 0$.

4) $\gamma = \hat{\gamma}_0$ *and* $\Delta = \hat{\Delta}$. **5)** $\gamma' = \hat{\gamma}'_0$ *and* $\Delta' = \hat{\Delta}'$.

6) $\gamma_n = \gamma + \Delta(n-1)$ *for* $1 \leq n \leq n_\phi$.

7) $\hat{\gamma}'_n = \hat{\gamma}'_0 + \hat{\Delta}' n = \hat{\gamma}' + \hat{\Delta}'(n-1)$ *for* $1 \leq n \leq n_{\hat{\phi}}$.

8) $t = \Delta - \gamma$. **9)** $s = \hat{\gamma}'_0 = \hat{\gamma}' - \hat{\Delta}'$.

10) $\delta = \dfrac{\Delta}{\Delta - \gamma}$. **11)** $\hat{\delta} = \dfrac{\hat{\Delta}'}{\hat{\gamma}'_0} = \dfrac{\hat{\Delta}'}{\hat{\gamma}' - \hat{\Delta}'}$.

12) $t_r = t_i = t$. **13)** $s_r = s_i = s$. **14)** $\delta_r = \delta_i = \delta$. **15)** $\hat{\delta}_r = \hat{\delta}_i = \hat{\delta}$.

16) *If* $\Delta = \Delta'$ *and* $\gamma = \gamma'$, *then* $t + s = \Delta$ *and* $1/\delta + 1/\hat{\delta} = 1$.

17) *In general* $1/\delta + 1/\hat{\delta} = 1$.

If $1 \leq n_{\hat{\phi}} \leq n_\phi$, *then* $t/\Delta + s/\hat{\Delta}' = 1$, $\Delta = \hat{\Delta}' \iff \gamma = \hat{\gamma}'_0$.

It is important to note that the scaling relations $t_r = t_i = t$ and $s_r = s_i = s$ are a fundamental identity, as these sets of critical exponents are defined in terms of $m(p,0)$ and $w(p,z(0))$, where $h = 0 \in \mathbb{R}$. The relation $1/\delta + 1/\hat{\delta} = 1$ is also a fundamental identity which follows from equation (2.24) and the definition of these critical exponents. The calculation of these scaling relations will serve as a consistency check of this mathematical framework.

Before we present the proof of Theorem 5, which is given in Section 3.2.2 below, we first demonstrate that the critical exponents of EMT satisfy the critical exponent scaling relations therein. This verification is essential, as there exists a binary composite medium which realizes the effective parameter of EMT [87]. Through our exploration of EMT, we will uncover aspects which illuminate general features of critical transport transitions exhibited by two-phase random media. These features will be discussed in detail in Section 3.3.

3.2.1 Effective Medium Theory

An EMT for the effective parameter problem may be constructed from dilute limits [39]. The EMT approximation for σ^* with percolation threshold p_c is given by [39]

$$p\,\frac{\sigma_2 - \sigma^*}{1 + p_c\,(\sigma_2/\sigma^* - 1)} + (1-p)\,\frac{\sigma_1 - \sigma^*}{1 + p_c\,(\sigma_1/\sigma^* - 1)} = 0. \tag{3.9}$$

Equation (3.9) leads to quadratic formulas involving $m(p,h) = \sigma^*/\sigma_2$ and $w(p,z(h)) = \sigma^*/\sigma_1$. The quadratic equation demonstrates that the relation $m(p,h) = h\,w(p,z(h))$ in (2.24) is exactly satisfied and that

$$m(p,h(s)) = \frac{-b\,(s,p,p_c) + \sqrt{-\zeta(s,p)}}{2s(1-p_c)}, \quad \zeta(\lambda,p) = -\lambda^2 + 2(1-\varphi)\lambda + \upsilon^2 - (1-\varphi)^2,$$
$$w(p,z(t)) = \frac{-b\,(s,1-p,p_c) + \sqrt{-\zeta(t,1-p)}}{2t(1-p_c)}, \quad \zeta(\lambda,1-p) = -\lambda^2 + 2\varphi\lambda + \upsilon^2 - \varphi^2, \tag{3.10}$$

where $b(\lambda,p,p_c) = (2p_c-1)\lambda + (1-p-p_c)$, $\varphi = \varphi(p,p_c) = p\,(1-p_c) + p_c(1-p)$, and $\upsilon = \upsilon(p,p_c) = 2\sqrt{p\,(1-p)\,p_c(1-p_c)}$.

The spectral measures μ and α in (2.13) may be extracted from equation (3.10) using the Stieltjes–Perron Inversion Theorem in (2.34). These measures are absolutely continuous, i.e., there exist density functions such that $\mu(d\lambda) = \mu(\lambda)d\lambda$ and $\alpha(d\lambda) = \alpha(\lambda)d\lambda$. Direct calculation shows that, for $p \neq p_c, 1-p_c$, these measures have gaps in the spectra about $\lambda = 0,1$: $\mu(\lambda) = 0 \iff \zeta(\lambda,p) \leq 0 \iff |\lambda-(1-\varphi)| \geq \upsilon$ and $\alpha(\lambda) = 0 \iff \zeta(\lambda,1-p) \leq 0 \iff |\lambda - \varphi| \geq \upsilon$. Therefore, the composite medium underlying the percolation model of EMT [87] is of class $\mathcal{B}_{n_\phi}$ and $\mathcal{B}_{n_{\hat{\phi}}}$ for $p \neq p_c, 1-p_c$ with $n_\phi, n_{\hat{\phi}} = \infty$ (see Definition 1). The Stieltjes transforms of μ and α are given by

$$F(p,s) = \int_{\lambda_0}^{1-\theta} \frac{\sqrt{\zeta(\lambda,p)}\,d\lambda}{2\pi(1-p_c)\,\lambda(s-\lambda)}\,, \qquad G(p,t) = \int_{\theta}^{\hat{\lambda}_1} \frac{\sqrt{\zeta(\lambda,1-p)}\,d\lambda}{2\pi(1-p_c)\,\lambda(t-\lambda)}\,, \tag{3.11}$$

where $\theta = \theta(p,p_c) = \varphi - \upsilon$ and $\hat{\lambda}_1 = 1 - \lambda_0 = \varphi + \upsilon$ define *spectral gaps*, which satisfy $\lim_{p\longrightarrow 1-p_c} \lambda_0 = 0$, $\lim_{p\longrightarrow p_c} \theta = 0$, and $\lim_{p\longrightarrow 1-p_c} \hat{\lambda}_1 = 1$.

Define a critical exponent Δ for the spectral gap $\theta(p) \sim |p-p_c|^\Delta$, as $p \longrightarrow p_c$, in $\mu(d\lambda)$ about $\lambda = 1$ and $\alpha(d\lambda)$ about $\lambda = 0$. Using the definition of a critical exponent in (3.4) and L'Hôpital's rule we have shown that $\Delta = 2$. Moreover $\lambda_0 = 1 - \hat{\lambda}_1 \sim |p-(1-p_c)|^\Delta$, as $p \longrightarrow 1-p_c$, with the same critical exponent. The absolutely continuous nature of the measures μ and α in EMT implies that critical indices are the same for $p \longrightarrow p_c^+$ and $p \longrightarrow p_c^-$. Therefore the spectral symmetry properties in the hypothesis of Lemma 13 hold for EMT.

We have explicitly calculated the integrals in equation (3.11) for real and complex h using the symbolic mathematics software Maple 15. Using the exact representation in (3.11) of $G(p,t(h))$, as a function of $0 \leq \theta \ll 1$ and $0 \leq |h| \ll 1$, we have calculated the critical exponents s, $\hat{\delta}$, $\hat{\delta}_r$, $\hat{\delta}_i$, and $\hat{\gamma}_n$, for $n = 0,1,2,\ldots$ These results are in agreement with our general theory. With $h = 0$ and $0 < \theta \ll 1$, we found that $w(p,z(0)) \sim \theta^{-1/2}$ which yields

$s = \Delta/2 = 1$. When $\theta = 0$ and $0 < h \ll 1$, one must split up the integration domain, $\Sigma_\alpha \supset (0, h-\epsilon) \cup (h+\epsilon, \hat{\lambda}_1)$, and take the principal value of the integral as $\epsilon \longrightarrow 0$. Doing so yields $\hat{\delta} = \hat{\delta}_r = \hat{\delta}_i = 2$. As in our general theory, the values of the exponents are independent of the path of h to zero. More specifically, these relations hold for $0 < |h_r| = |ah_i| \ll 1$ with arbitrary $a \in \mathbb{R}$, and for independent h_r and h_i satisfying $0 < |h_r|, |h_i| \ll 1$. The critical exponents $\hat{\gamma}_n$ associated with the moments $\hat{\phi}_n$ of the measure $\hat{\phi}$ satisfy our general relation $\hat{\gamma}_n = \hat{\gamma}_0 + \Delta n$ with $\hat{\gamma}_0 = \Delta = 2$ so that $\hat{\gamma}_n = \Delta(n+1)$.

Similarly, using the exact representation of $F(p, s(h))$ in (3.11), as a function of $0 \leq \theta \ll 1$ and $0 \leq |h| \ll 1$, we have calculated the critical exponents t, δ, δ_r, δ_i, and γ_n, for $n = 0, 1, 2, \ldots$ These results are also in agreement with our general theory. In accordance with [39], we obtain $t = \Delta/2 = 1$, so that the relation $s + t = \Delta = 2$ is satisfied. By direct calculation we have obtained $\delta = \delta_r = \delta_i = 2$. We have also obtained these values using $m(p, h) = hw(p, z(h))$ and the associated relations for complex h, $m_r = h_r w_r - h_i w_i$ and $m_i = h_r w_i + h_i w_r$, with $\hat{\delta} = \hat{\delta}_r = \hat{\delta}_i$ and $1/\delta + 1/\hat{\delta} = 1$. The mass $\phi_0(p) = F(p, 1)$ of the measure ϕ behaves logarithmically as $\theta \longrightarrow 0$, yielding $\gamma_0 = 0$. The exponents of the higher moments satisfy our general relation $\gamma_n = \gamma_0 + \Delta n = \gamma + \Delta(n-1)$, or $\gamma_n = \Delta n$, $n \geq 0$.

In summary, we have extended EMT to the complex quasi-static regime and shown that the critical exponents of EMT exactly satisfy our scaling relations displayed in Theorem 5. Moreover we have shown that, in EMT, the percolation threshold p_c and $1 - p_c$ coincide with the collapse of gaps in the spectral measures about the spectral endpoints $\lambda = 0, 1$. This is the behavior displayed in Lemma 2 and Corollary Corollary 1, which hold for general percolation models of stationary two-phase random media with $m(0) = m(p, 0)$ and $w(0) = w(p, 0)$. We will discuss this link between spectral gaps and the percolation threshold in more detail in Section 3.3.

3.2.2 Proof of Theorem 5

Baker's critical theory characterizes phase transitions of a given system via the asymptotic behavior of the underlying Stieltjes functions near a critical point. This powerful method has been very successful for the Ising model, precisely characterizing the phase transition (spontaneous magnetization) [4]. We will now show how this method may be adapted to provide a detailed description of phase transitions in transport, exhibited by binary composite media. Theorem 5 will be proven via a sequence of lemmas as we collect some important properties of $m(p, h)$, $g(p, h)$, $w(p, z(h))$, and $\hat{g}(p, h)$, and how they are related. We stress that the only assumption needed for Theorem 5 is that our percolation model is of class $\mathcal{B}_{n_\phi}$ and $\mathcal{B}_{n_{\hat{\phi}}}$ for $p > p_c$ and $p < p_c$, respectively, and $1 \le n_{\hat{\phi}} \le n_\phi$ (see Definition 1). The following theorem [4] characterizes Stieltjes functions (series of Stieltjes).

Theorem 6 *Let $D(i,j)$ denote the determinant*

$$D(i,j) = \begin{vmatrix} \xi_i & \xi_{i+1} & \cdots & \xi_{i+j} \\ \vdots & \vdots & \ddots & \vdots \\ \xi_{i+j} & \xi_{i+j+1} & \cdots & \xi_{i+2j} \end{vmatrix}. \tag{3.12}$$

The ξ_n form a series of Stieltjes if and only if $D(i,j) \ge 0$ for all $i, j = 0, 1, 2, \ldots$

Baker's inequalities for the sequences γ_n and $\hat{\gamma}_n$ of transport follow directly from Theorem 6. For example, $\phi_n \sim (p-p_c)^{-\gamma_n}$ and Theorem 6 with $\phi_i = \xi_i$, $i = n$, and $j = 1$, imply that, for $0 < p - p_c \ll 1$,

$$\begin{aligned} &(p-p_c)^{-\gamma_n-\gamma_{n+2}} - (p-p_c)^{-2\gamma_{n+1}} \ge 0 \iff (p-p_c)^{-\gamma_n-\gamma_{n+2}+2\gamma_{n+1}} \ge 1 \\ &\iff -\gamma_n - \gamma_{n+2} + 2\gamma_{n+1} \le 0 \iff \boxed{\gamma_{n+1} - 2\gamma_n + \gamma_{n-1} \ge 0}. \end{aligned} \tag{3.13}$$

The sequence of inequalities in (3.13) are *Baker's inequalities* for transport, corresponding to $m(p,h)$, and they imply that the sequence γ_n increases at least linearly with n. The symmetries in equations (2.18), (3.7), and (3.8) imply that Baker's inequalities also hold for the sequences $\hat{\gamma}'_n$, γ'_n, and $\hat{\gamma}_n$.

The following lemma provides the asymptotic behavior of the h derivatives of $g(p,h)$ and $\hat{g}(p,h)$, which will be used extensively in this section.

Lemma 3 *Let $h \in \mathcal{U}_0$ and $0 < |h| \ll 1$ and $|p - p_c| \ll 1$. Then the integrals in equation* (2.28) *have the following asymptotics for $n \ge 0$*

$$\left|\frac{\partial^n g(p,h)}{\partial h^n}\right| \sim \phi_n, \qquad \left|\frac{\partial^n \hat{g}(p,h)}{\partial h^n}\right| \sim \hat{\phi}_n. \tag{3.14}$$

Proof: The asymptotic behavior in equation (3.14) follows from equations (2.20), (2.21), (2.23), Baker's inequalities (3.13), and equation (2.18) ($g(p,h) = sF(p,s)$ and $\hat{g}(p,h) = -s\,G(p,t(s))$). These equations imply that, for $c_j, b_j \in \mathbb{Z}$ and $|p - p_c| \ll 1$,

$$\begin{aligned} \lim_{h\longrightarrow 0} \frac{\partial^n g(p,h)}{\partial h^n} &= \sum_{j=0}^{n} c_j \lim_{s\longrightarrow 1} \frac{\partial^j F(p,s)}{\partial s^j} \sim \phi_n\,, \\ \lim_{h\longrightarrow 0} \frac{\partial^n \hat{g}(p,h)}{\partial h^n} &= \sum_{j=0}^{n} b_j \lim_{s\longrightarrow 1} \frac{\partial^j G(p,t(s))}{\partial t^j} \sim \hat{\phi}_n \;\; \square. \end{aligned}$$

Lemma 3 demonstrates that the numbers n_ϕ and $n_{\hat{\phi}}$ introduced in Definition 1 are also related to the number of *finite* moments of the measures ϕ and $\hat{\phi}$, respectively.

Lemma 4 $\gamma_1 = \gamma$, $\gamma_1' = \gamma'$, $\hat{\gamma}_1 = \hat{\gamma}$, *and* $\hat{\gamma}_1' = \hat{\gamma}'$

Proof: Set $0 < p - p_c \ll 1$. By equations (2.18) ($g(p,h) = sF(p,s)$), (2.21), (3.7), and (3.13)

$$(p-p_c)^{-\gamma} \sim \chi(p,0) = \frac{\partial m(p,0)}{\partial h} = \lim_{s \longrightarrow 1} \left[-\frac{\partial F(p,s)}{\partial s} \right] = \phi_0 + \phi_1 \sim \phi_1 \sim (p-p_c)^{-\gamma_1}, \tag{3.15}$$

hence $\gamma_1 = \gamma$. Similarly for $0 < p_c - p \ll 1$, we have $\gamma_1' = \gamma'$. By equation (3.15), the symmetries between $m(p,h)$ and $w(p,z(h))$ given in (2.18), and the critical exponent definitions given in (3.7) and (3.8), we also have $\hat{\gamma}_1 = \hat{\gamma}$ and $\hat{\gamma}_1' = \hat{\gamma}'$ $\square$.

Equation (2.24) is consistent with, and provides a link between equations (3.5) and (3.6). We will see that the fundamental asymmetry between $m(p,h)$ and $w(p,z(h))$ ($\gamma_0' = 0$ and $\hat{\gamma}_0' > 0$), given in Theorem 5.2–3, is a direct and essential consequence of equation (2.24), and has deep and far reaching implications.

Lemma 5 *Let the sequences* γ_n *and* γ_n', $n \geq 0$, *be defined as in equation* (3.7). *Then*

1) $\gamma_0' = 0$, $\gamma_0 < 0$, $\gamma_n' > 0$, *and* $\gamma_n > 0$, *for* $n \geq 1$.

2) $0 < \lim_{h \longrightarrow 0} \langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle / E_0^2 < 1$ *for all* $p \in [0,1]$.

Proof: By equation (3.6) $|w(p,z(0))|$ is bounded for all $p < p_c$. Thus for all $p < p_c$, equations (2.21), (2.24), and (3.7) imply that

$$0 = \lim_{h \longrightarrow 0} hw(p,z(h)) = \lim_{h \longrightarrow 0} m(p,h) = \lim_{s \longrightarrow 1} (1 - F(p,s)) = 1 - \phi_0(p) \sim 1 - (p_c - p)^{-\gamma_0'},$$

where the rightmost relation holds for $0 < p_c - p \ll 1$ and the leftmost relation is consistent with equation (3.5). Therefore, $\gamma_0' = 0$ and ϕ is a probability measure for all $p < p_c$. The strict positivity of the γ_n', for $n \geq 1$, follows from Baker's inequalities in (3.13). Thus, from the analogy of equation (3.15) for $p < p_c$, we have

$$\infty = \lim_{p \longrightarrow p_c^-} \phi_1(p) = - \lim_{p \longrightarrow p_c^-} \frac{\partial m(p,0)}{\partial h} \, . \tag{3.16}$$

For $p > p_c$, equations (2.21) and (3.5) imply that $0 < \lim_{h \longrightarrow 0} |m(p,h)| = 1 - \phi_0 < 1$. Therefore, $(p-p_c)^{-\gamma_0} \sim \phi_0 < 1$ for all $0 < p - p_c \ll 1$, hence $\gamma_0 < 0$. The strict positivity of γ_1 follows from equation (3.16), and the strict positivity of the γ_n for $n \geq 2$ follows from Baker's inequalities (3.13). Equation (2.22) and the inequality $0 < \lim_{h \longrightarrow 0} |m(p,h)| = 1 - \phi_0 < 1$ imply that $0 < \lim_{h \longrightarrow 0} \langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle / E_0^2 < 1$ for all $p \in [0,1]$.

Lemma 6 *Let the sequence* $\hat{\gamma}'_n$, $n \geq 0$, *be defined as in equation* (3.8). *Then*

$$\mathbf{1)} \quad \hat{\gamma}'_n > 0 \ \ \textit{for all} \ \ n \geq 0. \qquad\qquad \mathbf{2)} \quad \lim_{p \longrightarrow p_c, h \longrightarrow 0} \langle E_f^2 \rangle = \infty.$$

Proof: By equation (3.5) we have $0 < \lim_{h \longrightarrow 0} |m(p,h)| < 1$, for all $p > p_c$. Therefore equation (2.24) implies that $\lim_{h \longrightarrow 0} w(p, z(h)) = \lim_{h \longrightarrow 0} m(p,h)/h = \infty$, for all $p > p_c$, which is consistent with equation (3.6). More specifically, for all $p > p_c$, equations (2.24) and (3.5) imply that $0 < \lim_{h \longrightarrow 0} |m(p,h)| = \lim_{h \longrightarrow 0} |hw(p, z(h))| = L(p) < 1$, where $L(p) = 0$ for all $p < p_c$. Therefore, by equation (2.18), we have

$$\begin{aligned} &\lim_{h \longrightarrow 0} |h\, w(p, z(h))| = \lim_{h \longrightarrow 0} |h\, \hat{g}(p,h)| \in (0,1), \ \text{for all } p > p_c, \\ &\lim_{h \longrightarrow 0} |h\, w(p, z(h))| = \lim_{h \longrightarrow 0} |h\, \hat{g}(p,h)| = 0, \ \text{for all } p < p_c\,. \end{aligned} \tag{3.17}$$

By equations (2.23), (3.6), and (3.8) we have, for all $p > p_c$,

$$\begin{aligned} \infty = \lim_{p \longrightarrow p_c^-} \lim_{h \longrightarrow 0} w(p, z(h)) &= \lim_{p \longrightarrow p_c^-} \lim_{s \longrightarrow 1} (1 - G(p, t(s))) = 1 + \lim_{p \longrightarrow p_c^-} \hat{\phi}_0(p) \\ \sim 1 + \lim_{p \longrightarrow p_c^-} (p_c - p)^{-\hat{\gamma}'_0}, \end{aligned}$$

hence $\hat{\gamma}'_0 > 0$. Baker's inequalities then imply that $\hat{\gamma}'_n > 0$ for all $n \geq 0$. Equation (2.22) and the analogy thereof involving $\hat{\phi}_1$, and the strict positivity of $\hat{\gamma}'_1$, γ_1, and γ'_1, shown here and in Lemma 5, imply that $\lim_{p \longrightarrow p_c, h \longrightarrow 0} \langle E_f^2 \rangle = \infty$ $\square$.

The asymptotic behavior of $|\hat{g}(p,h)|$ in equation (3.14), and Lemma 6 motivates the following fundamental homogenization assumption of this section [4]:

Remark 2 *Near the critical point* $(p,h) = (p_c, 0)$, *the asymptotic behavior of the Stieltjes function* $\hat{g}(p,h)$ *is determined primarily by the mass* $\hat{\phi}_0(p)$ *of the measure* $\hat{\phi}$ *and the rate of collapse of the spectral gap* θ_α.

By Remark 2, and in light of Lemmas 4–6, we make the following variable changes:

$$\begin{aligned} &\hat{q} = y(p_c - p)^{\hat{\Delta}'}, \qquad \hat{Q}(p) = \hat{S}(p)(p_c - p)^{\hat{\Delta}'}, \qquad d\hat{\pi}(\hat{q}) = (p_c - p)^{\hat{\gamma}'_0}\, d\hat{\phi}(y), \\ &q = y(p - p_c)^{\Delta}, \qquad Q(p) = S(p)(p - p_c)^{\Delta}, \qquad d\pi(q) = (p - p_c)^{\gamma}\, y\, d\phi(y), \end{aligned} \tag{3.18}$$

so that, by equations (3.7) and (3.8), $\hat{Q}(p), Q(p) \sim 1$ and the masses $\hat{\pi}_0$ and π_0 of the measures $\hat{\pi}$ and π, respectively, satisfy $\hat{\pi}_0, \pi_0 \sim 1$ as $p \longrightarrow p_c$.

Equation (3.18) defines the following scaling functions $G_{n-1}(x)$, $\hat{G}_n(\hat{x})$, $\mathcal{G}_{n-1,j}(x)$, and $\hat{\mathcal{G}}_{n,j}(\hat{x})$ as follows. For $h \in \mathcal{U}_0 \cap \mathbb{R}$, equations (2.28) and (3.18) imply, for all $n \geq 1$, that

$$\begin{aligned}
&\frac{\partial^n g}{\partial h^n} \propto (p-p_c)^{-(\gamma+\Delta(n-1))} G_{n-1}(x), && \frac{\partial^n \hat{g}}{\partial h^n} \propto (p_c-p)^{-(\hat{\gamma}_0'+\hat{\Delta}' n)} \hat{G}_n(\hat{x}), \\
&G_{n-1}(x) = \int_0^{Q(p)} \frac{q^{n-1} d\pi(q)}{(1+xq)^{n+1}}, && \hat{G}_n(\hat{x}) = \int_0^{\hat{Q}(p)} \frac{\hat{q}^{\,n} d\hat{\pi}(\hat{q})}{(1+\hat{x}\hat{q})^{n+1}}, \\
&x = h(p-p_c)^{-\Delta}, \quad 0 < p-p_c \ll 1, && \hat{x} = h(p_c-p)^{-\hat{\Delta}'}, \quad 0 < p_c - p \ll 1,
\end{aligned} \tag{3.19}$$

respectively, and an analogue of (3.19) for the opposite limits involving $\hat{\Delta}$, $\hat{\gamma}_0$, Δ', and γ'. For $h \in \mathcal{U}_0$ such that $h_i \neq 0$, we define the scaling functions $\mathcal{R}_{n-1}(x)$, $\mathcal{I}_{n-1}(x)$, $\hat{\mathcal{R}}_n(\hat{x})$, and $\hat{\mathcal{I}}_n(\hat{x})$ as follows. Using equations (2.30) and (3.18) we have, for $0 < p - p_c \ll 1$,

$$\begin{aligned}
\frac{\partial^n g}{\partial h^n} &= (-1)^n n! \sum_{j=0}^{n+1} \binom{n+1}{j} \bar{h}^j \int_0^{S(p)} \frac{y^{n+j} d\phi(y)}{|1+hy|^{2(n+1)}} \\
&= (-1)^n n! \sum_{j=0}^{n+1} \binom{n+1}{j} [\bar{x}(p-p_c)^{\Delta}]^j (p-p_c)^{-(\gamma+\Delta(n-1+j))} \mathcal{G}_{n-1,j}(x) \\
&= (-1)^n n! (p-p_c)^{-(\gamma+\Delta(n-1))} \mathcal{K}_{n-1}(x), \quad \mathcal{K}_{n-1}(x) = \mathcal{R}_{n-1}(x) + \mathrm{i}\,\mathcal{I}_{n-1}(x), \\
\frac{\partial^n \hat{g}}{\partial h^n} &= (-1)^n n! (p-p_c)^{-(\hat{\gamma}_0+\hat{\Delta} n)} \hat{\mathcal{K}}_n(\hat{x}), \quad \hat{\mathcal{K}}_n(\hat{x}) = \hat{\mathcal{R}}_n(\hat{x}) + \mathrm{i}\,\hat{\mathcal{I}}_n(\hat{x}).
\end{aligned} \tag{3.20}$$

Here, x and $\hat{x}$ are defined in equation (3.19) and

$$\begin{aligned}
&\mathcal{G}_{n-1,j}(x) = \int_0^{Q(p)} \frac{q^{n-1+j} d\pi(q)}{|1+xq|^{2(n+1)}}, && \hat{\mathcal{G}}_{n,j}(\hat{x}) = \int_0^{\hat{Q}(p)} \frac{\hat{q}^{\,n+j} d\hat{\pi}(\hat{q})}{|1+\hat{x}\hat{q}|^{2(n+1)}}, \\
&\mathcal{K}_{n-1}(x) = \sum_{j=0}^{n+1} \binom{n+1}{j} \bar{x}^{\,j} \mathcal{G}_{n-1,j}(x), && \hat{\mathcal{K}}_n(\hat{x}) = \sum_{j=0}^{n+1} \binom{n+1}{j} \bar{\hat{x}}^j \hat{\mathcal{G}}_{n,j}(\hat{x}),
\end{aligned} \tag{3.21}$$

where we have made the definitions $\mathcal{R}_{n-1}(x) = \mathrm{Re}\,\mathcal{K}_{n-1}(x)$, $\mathcal{I}_{n-1}(x) = \mathrm{Im}\,\mathcal{K}_{n-1}(x)$, $\hat{\mathcal{R}}_n(\hat{x}) = \mathrm{Re}\,\hat{\mathcal{K}}_n(\hat{x})$, and $\hat{\mathcal{I}}_n(\hat{x}) = \mathrm{Im}\,\hat{\mathcal{K}}_n(\hat{x})$. Analogous formulas are defined for the opposite limit, $0 < p_c - p \ll 1$, involving $\hat{\Delta}'$, $\hat{\gamma}_0'$, Δ', and γ'. By (2.19) we have, for $p \in [0,1]$ and $n \geq 0$,

$$G_{n-1}(x) > 0, \quad \mathcal{G}_{n-1,j}(x) > 0, \qquad \hat{G}_n(\hat{x}) > 0, \quad \hat{\mathcal{G}}_{n,j}(\hat{x}) > 0. \tag{3.22}$$

By hypothesis, our percolation model is of class $\mathcal{B}_{n_\phi}$ and $\mathcal{B}_{n_{\hat{\phi}}}$ for every $p > p_c$ and $p < p_c$, respectively, and $1 \leq n_\phi \leq n_{\hat{\phi}}$ (see Definition 1). We therefore have

$$\begin{aligned}
&\lim_{h \longrightarrow 0} G_{n-1}(x) < \infty, && \lim_{h \longrightarrow 0} \mathcal{G}_{n-1,j}(x) < \infty, && \text{for all} \ \ p > p_c, \quad 0 \leq n \leq n_\phi \\
&\lim_{h \longrightarrow 0} \hat{G}_n(\hat{x}) < \infty, && \lim_{h \longrightarrow 0} \hat{\mathcal{G}}_{n,j}(\hat{x}) < \infty, && \text{for all} \ \ p < p_c, \quad 0 \leq n \leq n_{\hat{\phi}}.
\end{aligned} \tag{3.23}$$

Lemma 7 *Let $\hat{G}_n(\hat{x})$, $G_{n-1}(x)$, and the associated critical exponents be defined as in equation* (3.19)*, for $p > p_c$. Then, if $n_\phi \geq 1$,*

1) $G_{n-1}(x) \sim 1$ *as* $x \longrightarrow 0$ $(h \longrightarrow 0$ *and* $0 < p - p_c \ll 1)$ *for all* $1 \leq n \leq n_\phi$.

2) $[\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})] \sim 1$ *as* $\hat{x} \longrightarrow 0$ $(h \longrightarrow 0$ *and* $0 < p - p_c \ll 1)$
for all $1 \leq n \leq n_{\hat{\phi}}$.

3) $\gamma = \hat{\gamma}_0\,.$ **4**) $\Delta = \hat{\Delta}\,.$

Proof: oLet $h \in \mathcal{U}_0 \cap \mathbb{R}$ and $p > p_c$. Equations (2.29), (3.19), (3.22), and (3.23) imply that we have, for all $1 \leq n \leq n_\phi$, $0 < p - p_c \ll 1$, and $0 < h \ll 1$,

$$(0,\infty) \ni (p-p_c)^{-(\gamma+\Delta(n-1))}G_{n-1}(x) = (p-p_c)^{-(\hat{\gamma}_0+\hat{\Delta}(n-1))}[\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})]. \quad (3.24)$$

Equations (3.22) and (3.23) imply that $G_{n-1}(x) \sim 1$ as $x \longrightarrow 0$, for all $1 \leq n \leq n_\phi$. Equation (3.24) then implies that $[\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})] \sim 1$ as $\hat{x} \longrightarrow 0$, for all $1 \leq n \leq n_\phi$ (a possible competition in sign between two diverging terms). Or equivalently, generalizing equation (3.17), $[\hat{G}_0(\hat{x}) - \hat{x}^n\hat{G}_n(\hat{x})] \sim 1$. Therefore, equation (3.24) implies that

$$\gamma + \Delta(n-1) = \hat{\gamma}_0 + \hat{\Delta}(n-1), \quad 1 \leq n \leq n_\phi, \quad (3.25)$$

which in turn, implies that $\gamma = \hat{\gamma}_0$ and $\Delta = \hat{\Delta}$ □.

Lemma 8 *Let $\hat{G}_n(\hat{x})$, $G_{n-1}(x)$, and the associated critical exponents be defined as in equation* (3.19)*, for $p < p_c$. Then, if $n_{\hat{\phi}} \geq 1$,*

1) $[\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})] \sim 1$ *as* $\hat{x} \longrightarrow 0$ $(h \longrightarrow 0$ *and* $0 < p_c - p \ll 1)$,
for all $1 \leq n \leq n_{\hat{\phi}}$.

2) $G_{n-1}(x) \sim 1$ *as* $x \longrightarrow 0$ $(h \longrightarrow 0$ *and* $0 < p_c - p \ll 1$, *for all* $1 \leq n \leq n_{\hat{\phi}}$.

3) $\gamma' = \hat{\gamma}'_0$. **4**) $\Delta' = \hat{\Delta}'$.

Proof: Let $h \in \mathcal{U}_0 \cap \mathbb{R}$ and $p < p_c$. Equations (2.29), (3.19), (3.22), and (3.23) imply that, for all $1 \leq n \leq n_{\hat{\phi}}$, $0 < p_c - p \ll 1$, and $0 < h \ll 1$,

$$(0,\infty) \ni (p_c-p)^{-(\hat{\gamma}'_0+\hat{\Delta}'(n-1))}[\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})] = (p_c-p)^{-(\gamma'+\Delta'(n-1))}G_{n-1}(x) \quad (3.26)$$

Equations (3.22) and (3.23) imply that $[\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})] \sim 1$ as $\hat{x} \longrightarrow 0$ for every $1 \leq n \leq n_{\hat{\phi}}$. Equation (3.26) then implies that $G_{n-1}(x) \sim 1$ as $x \longrightarrow 0$ for all $1 \leq n \leq n_{\hat{\phi}}$. Therefore,

$$\gamma' + \Delta'(n-1) = \hat{\gamma}'_0 + \hat{\Delta}'(n-1), \quad 1 \leq n \leq n_{\hat{\phi}}.$$

Which in turn, implies that $\gamma' = \hat{\gamma}'_0$ and $\Delta' = \hat{\Delta}'$ □.

Lemma 9 *Let $\hat{G}_n(\hat{x})$, $G_{n-1}(x)$, and the associated critical exponents be defined as in equation* (3.19)*. Then, if* $1 \le n_{\hat{\phi}} \le n_\phi$,

1) $\gamma_n = \gamma + \Delta(n-1)$, *for all* $1 \le n \le n_\phi$.

2) $\hat{\gamma}'_n = \hat{\gamma}'_0 + \hat{\Delta}' n = \hat{\gamma}' + \hat{\Delta}'(n-1)$, *for all* $1 \le n \le n_{\hat{\phi}}$.

3) $t = \hat{\Delta} - \hat{\gamma}_0 = \Delta - \gamma$.

4) $s = \hat{\gamma}'_0 = \hat{\gamma}' - \hat{\Delta}'$.

Proof: Let $0 < p - p_c \ll 1$. By equations (3.7), (3.14), and (3.19), and Lemma 7 we have, for all $1 \le n \le n_\phi$,

$$\begin{aligned}(p-p_c)^{-\gamma_n} \sim \phi_n \sim \lim_{h\longrightarrow 0} \frac{\partial^n g(p,h)}{\partial h^n} &\sim (p-p_c)^{-(\gamma+\Delta(n-1))} \lim_{x\longrightarrow 0} G_{n-1}(x) \\ &\sim (p-p_c)^{-(\gamma+\Delta(n-1))}.\end{aligned}$$

Therefore $\gamma_n = \gamma + \Delta(n-1)$ for all $1 \le n \le n_\phi$, with constant gap $\gamma_i - \gamma_{i-1} = \Delta$. When this is true for all $n \ge 0$, as in EMT where $n_\phi = \infty$, this is consistent with the absence of multifractal behavior for the bulk conductivity $\sigma^*(p,h) = \sigma_2 m(p,h)$ [116].

Now let $0 < p_c - p \ll 1$. By equations (3.8), (3.14), (3.19), (3.22) and (3.23) we have, for all $1 \le n \le n_{\hat{\phi}}$,

$$(p_c-p)^{-\hat{\gamma}_n} \sim \hat{\phi}_n \sim \lim_{h\longrightarrow 0} \frac{\partial^n \hat{g}(p,h)}{\partial h^n} \propto (p_c-p)^{-(\hat{\gamma}'_0+\hat{\Delta}' n)} \lim_{\hat{x}\longrightarrow 0} \hat{G}_n(\hat{x}) \sim (p_c-p)^{-(\hat{\gamma}'_0+\hat{\Delta}' n)}.$$

Therefore, by Lemma 4, we have $\hat{\gamma}_n = \hat{\gamma}'_0 + \hat{\Delta}' n = \hat{\gamma}' + \hat{\Delta}'(n-1)$ for all $1 \le n \le n_{\hat{\phi}}$, with constant gap $\hat{\gamma}'_i - \hat{\gamma}'_{i-1} = \hat{\Delta}$. When this is true for all $n \ge 0$, as in EMT where $n_{\hat{\phi}} = \infty$, this is consistent with the absence of multifractal behavior for the bulk conductivity $\sigma^*(p,z(h)) = \sigma_1 w(p,z(h))$ [116].

Again let $0 < p - p_c \ll 1$. Equations (2.18), (2.25), (3.7), (3.17), and (3.19) yield

$$\begin{aligned}(p-p_c)^t \sim \lim_{h\longrightarrow 0} m(p,h) = 1 - \lim_{h\longrightarrow 0} g(p,h) = \lim_{h\longrightarrow 0} h\hat{g}(p,h) &= (p-p_c)^{\hat{\Delta}-\hat{\gamma}_0} \lim_{\hat{x}\longrightarrow 0} \hat{x}\hat{G}_0(\hat{x}) \\ \sim (p-p_c)^{\hat{\Delta}-\hat{\gamma}_0}.\end{aligned} \tag{3.27}$$

Therefore, by Lemma 7 we have, for $n_\phi \ge 1$, $t = \hat{\Delta} - \hat{\gamma}_0 = \Delta - \gamma$.

Finally let $0 < p_c - p \ll 1$. By equations (2.18), (3.8), (3.19), (3.22), and (3.23), and Lemma 6, we have

$$\begin{aligned}(p_c-p)^{-s} \sim \lim_{h\longrightarrow 0} w(p,z(h)) \sim 1 + \lim_{h\longrightarrow 0} \hat{g}(p,h) &= 1 + (p_c-p)^{-\hat{\gamma}'_0} \lim_{\hat{x}\longrightarrow 0} \hat{G}_0(\hat{x}) \\ \sim (p_c-p)^{-\hat{\gamma}'_0}.\end{aligned}$$

Therefore, by Lemma 9.2 we have, for $n_{\hat{\phi}} \ge 1$, $s = \hat{\gamma}'_0 = \hat{\gamma}' - \hat{\Delta}'$ $\square$.

Lemma 10 *Let $\hat{G}_n(\hat{x})$, $G_{n-1}(x)$, and the associated critical exponents be defined as in equation* (3.19)*, for $p > p_c$ and $p < p_c$. Then for all $1 \le n \le n_{\hat{\phi}} \le n_\phi$*

1) $G_{n-1}(x) \sim [\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})] \sim x^{-(\gamma+\Delta(n-1))/\Delta}$,

as $\hat{x} \longrightarrow \infty$ ($p \longrightarrow p_c^+$ *and* $0 < h \ll 1$) .

2) $G_{n-1}(x) \sim [\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})] \sim x^{-(\gamma'+\Delta'(n-1))/\Delta'}$,

as $x \longrightarrow \infty$ ($p \longrightarrow p_c^-$ *and* $0 < h \ll 1$) .

3) $\delta = \hat{\Delta}/(\hat{\Delta} - \hat{\gamma}_0) = \Delta/(\Delta - \gamma)$.

4) $\hat{\delta} = \hat{\Delta}'/\hat{\gamma}_0' = \hat{\Delta}'/(\hat{\gamma}' - \hat{\Delta}')$.

Proof: Let $0 < h \ll 1$, so that $g(p,h)$ and $\hat{g}(p,h)$ are analytic for all $p \in [0,1]$ [65]. The analyticity of $g(p,h)$ and $\hat{g}(p,h)$ implies that all orders of h derivatives of these functions are bounded as $p \longrightarrow p_c$, from the left or the right. Therefore, equation (3.24) holds for $0 < p - p_c \ll 1$, and equation (3.26) holds for $0 < p_c - p \ll 1$. Moreover, in order to cancel the diverging p dependent prefactors in (3.24) and (3.26) we must have, *in general*, for all $n \ge 1$,

$$G_{n-1}(x) \sim x^{-(\gamma+\Delta(n-1))/\Delta}, \quad [\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})] \sim \hat{x}^{-(\hat{\gamma}_0+\hat{\Delta}(n-1))/\hat{\Delta}}, \quad \text{as} \quad p \longrightarrow p_c^+,$$
$$G_{n-1}(x) \sim x^{-(\gamma'+\Delta'(n-1))/\Delta'}, \quad [\hat{G}_{n-1}(\hat{x}) - \hat{x}\hat{G}_n(\hat{x})] \sim \hat{x}^{-(\hat{\gamma}_0'+\hat{\Delta}'(n-1))/\hat{\Delta}'}, \quad \text{as} \quad p \longrightarrow p_c^-. \tag{3.28}$$

For $1 \le n \le n_{\hat{\phi}} \le n_\phi$, lemma 10.1 and 10.2 follow from (3.28) and Lemmas 7 and 8.

Now by equations (2.18), (2.24), (3.7), (3.19), and (3.28) for $n = 1$, we have

$$h^{1/\delta} \sim \lim_{p \longrightarrow p_c^+} m(p,h) = \lim_{p \longrightarrow p_c^+} hw(p,z(h)) \sim \lim_{p \longrightarrow p_c^+} h\hat{g}(p,h) = h \lim_{p \longrightarrow p_c^+} (p-p_c)^{-\hat{\gamma}_0}\hat{G}_0(\hat{x})$$
$$\sim h(p-p_c)^{-\hat{\gamma}_0} h^{-\hat{\gamma}_0/\hat{\Delta}} (p-p_c)^{-\hat{\Delta}(-\hat{\gamma}_0/\hat{\Delta})} = h^{(\hat{\Delta}-\hat{\gamma}_0)/\hat{\Delta}}. \tag{3.29}$$

Therefore $\delta = \hat{\Delta}/(\hat{\Delta} - \hat{\gamma}_0)$, and for $1 \le n_{\hat{\phi}} \le n_\phi$, Lemma 7 implies $\delta = \Delta/(\Delta - \gamma)$. Similarly by equations (2.18), (3.8), (3.19), and (3.28) for $n = 1$, and Lemma 6, we have

$$h^{-1/\hat{\delta}} \sim \lim_{p \longrightarrow p_c^-} w(p,z(h)) \sim 1 + \lim_{p \longrightarrow p_c^-} \hat{g}(p,h) = 1 + \lim_{p \longrightarrow p_c^-} (p-p_c)^{-\hat{\gamma}_0'}\hat{G}_0(\hat{x}) = h^{-\hat{\gamma}_0'/\hat{\Delta}'}, \tag{3.30}$$

in general. By Lemma 9, for $1 \le n_{\hat{\phi}} \le n_\phi$, we have $\hat{\delta} = \hat{\Delta}'/\hat{\gamma}_0' = \hat{\Delta}'/(\hat{\gamma}' - \hat{\Delta}')$ □.

Lemma 11 *Let $h \in \mathcal{U}_0$ such that $h_i \neq 0$, and $\hat{\mathcal{G}}_{n,j}(\hat{x})$, $\hat{\mathcal{R}}_n(\hat{x})$, $\hat{\mathcal{I}}_n(\hat{x})$, and the associated critical exponents be defined as in equations* (3.20) *and* (3.21) *for $p > p_c$ and $p < p_c$. Furthermore, let s_r, s_i, t_r, and t_i be defined as in equations* (3.7) *and* (3.8). *Then,*

1) $[\hat{\mathcal{G}}_{0,0}(\hat{x}) + \hat{x}_r\hat{\mathcal{G}}_{0,1}(\hat{x})] \sim \hat{x}_i\hat{\mathcal{G}}_{0,1}(\hat{x}) \sim 1$ *as* $\hat{x} \longrightarrow 0$ $(h \longrightarrow 0$ *and* $0 < p_c - p \ll 1)$.

2) $\lim_{\hat{x}\longrightarrow 0}[\hat{x}_r\hat{\mathcal{G}}_{0,0}(\hat{x}) + |\hat{x}|^2\hat{\mathcal{G}}_{0,1}(\hat{x})] \sim \lim_{\hat{x}\longrightarrow 0}[\hat{x}_i\hat{\mathcal{G}}_{0,0}(\hat{x})] \sim 1$ *for* $0 < p - p_c \ll 1$.

3) $s_r = s_i = \hat{\gamma}_0' = s$.

4) $t_r = t_i = \Delta - \gamma = t$.

Proof: Let $0 < p_c - p \ll 1$, $h \in \mathcal{U}_0$ such that $h_i \neq 0$, and $0 < |h| \ll 1$. By equations (3.20) and (3.21), for $n = 0$, we have

$$\hat{g}(p,h) = \int_0^{\hat{S}(p)} \frac{d\hat{\phi}(y)}{|1+hy|^2} + \bar{h}\int_0^{\hat{S}(p)} \frac{y\,d\hat{\phi}(y)}{|1+hy|^2} = (p_c - p)^{-\hat{\gamma}_0'}[\hat{\mathcal{G}}_{0,0}(\hat{x}) + \bar{\hat{x}}\hat{\mathcal{G}}_{0,1}(\hat{x})], \tag{3.31}$$

so that

$$\begin{aligned} \hat{g}_r(p,h) &= (p_c - p)^{-\hat{\gamma}_0'}\hat{\mathcal{R}}_0(\hat{x}) = (p_c - p)^{-\hat{\gamma}_0'}[\hat{\mathcal{G}}_{0,0}(\hat{x}) + \hat{x}_r\hat{\mathcal{G}}_{0,1}(\hat{x})] \\ \hat{g}_i(p,h) &= (p_c - p)^{-\hat{\gamma}_0'}\hat{\mathcal{I}}_0(\hat{x}) = -(p_c - p)^{-\hat{\gamma}_0'}\hat{x}_i\hat{\mathcal{G}}_{0,1}(\hat{x}). \end{aligned} \tag{3.32}$$

Equations (3.17) and (3.22) imply that $\hat{\mathcal{R}}_0(\hat{x}) \sim \hat{\mathcal{I}}_0(\hat{x}) \sim 1$ as $\hat{x} \longrightarrow 0$ ($h \longrightarrow 0$ and $0 < p_c - p \ll 1$). Therefore, equations (2.18), (3.8), (3.32) and Lemma 6 imply that

$$\begin{aligned} (p_c - p)^{-s_r} &\sim w_r(p, z(0)) \sim 1 + \hat{g}_r(p,0) \sim 1 + (p_c - p)^{-\hat{\gamma}_0'} \lim_{\hat{x}\longrightarrow 0}\hat{\mathcal{R}}_0(\hat{x}) \sim (p_c - p)^{-\hat{\gamma}_0'}, \\ (p_c - p)^{-s_i} &\sim w_i(p, z(0)) \sim \hat{g}_i(p,0) \sim (p_c - p)^{-\hat{\gamma}_0'} \lim_{\hat{x}\longrightarrow 0}\hat{\mathcal{I}}_0(\hat{x}) \sim (p_c - p)^{-\hat{\gamma}_0'}. \end{aligned} \tag{3.33}$$

Equation (3.33) and Lemma 9 imply that $s_r = s_i = \hat{\gamma}_0' = s$.

Now let $0 < p - p_c \ll 1$ with h as before. In equation (3.27) we demonstrated that $m(p,0) = \lim_{h\longrightarrow 0} h\hat{g}(p,h)$. Therefore equation (3.32), for $p > p_c$, implies that

$$\begin{aligned} m_r(p,0) &\sim \lim_{h\longrightarrow 0}[h_r\hat{g}_r(p,h) - h_i\hat{g}_i(p,h)] = (p - p_c)^{\hat{\Delta}-\hat{\gamma}_0} \lim_{\hat{x}\longrightarrow 0}[\hat{x}_r\hat{\mathcal{G}}_{0,0}(\hat{x}) + |\hat{x}_r|^2\hat{\mathcal{G}}_{0,1}(\hat{x})] \\ m_i(p,0) &\sim \lim_{h\longrightarrow 0}[h_i\hat{g}_r(p,h) + h_r\hat{g}_i(p,h)] = (p - p_c)^{\hat{\Delta}-\hat{\gamma}_0} \lim_{\hat{x}\longrightarrow 0}[\hat{x}_i\hat{\mathcal{G}}_{0,0}(\hat{x})] \end{aligned} \tag{3.34}$$

By equation (3.17) we have $\lim_{\hat{x}\longrightarrow 0}[\hat{x}_r\hat{\mathcal{G}}_{0,0}(\hat{x}) + |\hat{x}|^2\hat{\mathcal{G}}_{0,1}(\hat{x})] \sim \lim_{\hat{x}\longrightarrow 0}[\hat{x}_i\hat{\mathcal{G}}_{0,0}(\hat{x})] \sim 1$ for all $0 < p - p_c \ll 1$. Therefore, equations (3.7) and (3.34) imply that

$$(p - p_c)^{t_r} \sim m_r(p,0) \sim (p - p_c)^{\hat{\Delta}-\hat{\gamma}_0}, \qquad (p - p_c)^{t_i} \sim m_i(p,0) \sim (p - p_c)^{\hat{\Delta}-\hat{\gamma}_0}. \tag{3.35}$$

Equation (3.35) and Lemmas 7 and 9 imply that, for $1 \leq n_{\hat{\phi}} \leq n_\phi$, $t_r = t_i = \hat{\Delta} - \hat{\gamma}_0 = \Delta - \gamma = t$ $\square$.

Lemma 12 *Let $h \in \mathcal{U}_0$ such that $h_i \neq 0$, and $\hat{\mathcal{G}}_{n,j}(\hat{x})$, $\hat{\mathcal{R}}_n(\hat{x})$, $\hat{\mathcal{I}}_n(\hat{x})$, and the associated critical exponents be defined as in equations* (3.20) *and* (3.21) *for $p > p_c$ and $p < p_c$. Furthermore, let $\hat{\delta}_r$, $\hat{\delta}_i$, δ_r, and δ_i be defined as in equations* (3.7) *and* (3.8)*. Then,*

1) $\hat{\mathcal{R}}_0(\hat{x}) \sim \hat{\mathcal{I}}_0(\hat{x}) \sim |\hat{x}|^{-\hat{\gamma}_0'/\hat{\Delta}'}$, *as* $\hat{x} \longrightarrow \infty$ ($p \longrightarrow p_c^-$ *and* $0 < |h| \ll 1$).

2) $[\hat{x}_r \hat{\mathcal{R}}_0(\hat{x}) - \hat{x}_i \hat{\mathcal{I}}_0(\hat{x})] \sim [\hat{x}_r \hat{\mathcal{I}}_0(\hat{x}) + \hat{x}_i \hat{\mathcal{R}}_0(\hat{x})] \sim |\hat{x}|^{(\hat{\Delta}-\hat{\gamma}_0)/\hat{\Delta}}$, *as* $\hat{x} \longrightarrow \infty$.

3) $\hat{\delta}_r = \hat{\delta}_i = \hat{\Delta}'/\hat{\gamma}_0' = \hat{\delta}$.

4) $\delta_r = \delta_i = \Delta/(\Delta - \gamma) = \delta$.

Proof: Let $h \in \mathcal{U}_0$ such that $h_i \neq 0$ and $0 < |h| \ll 1$, so that $g(p,h)$ and $\hat{g}(p,h)$ are analytic for all $p \in [0,1]$ [65]. Equations (2.18), (3.8), (3.32) and Lemma 6 imply that

$$|h|^{-1/\hat{\delta}_r} \sim w_r(p_c, z(h)) \sim 1 + \hat{g}_r(p_c, h) \sim \lim_{p \longrightarrow p_c^-} (p_c - p)^{-\hat{\gamma}_0'} \hat{\mathcal{R}}_0(\hat{x}), \tag{3.36}$$
$$|h|^{-1/\hat{\delta}_i} \sim w_i(p_c, z(h)) \sim \hat{g}_i(p_c, h) \sim \lim_{p \longrightarrow p_c^-} (p_c - p)^{-\hat{\gamma}_0'} \hat{\mathcal{I}}_0(\hat{x}).$$

The analyticity of $g(p,h)$ and $\hat{g}(p,h)$ implies that they are bounded for all $p \in [0,1]$. Therefore, in order to cancel the diverging p dependent prefactors in equations (3.36), we must have $\hat{\mathcal{R}}_0(\hat{x}) \sim \hat{\mathcal{I}}_0(\hat{x}) \sim |x|^{-\hat{\gamma}_0'/\hat{\Delta}'}$ as $\hat{x} \longrightarrow \infty$ ($p \longrightarrow p_c^-$ and $0 < h \ll 1$). Equation (3.36) then implies

$$|h|^{-1/\hat{\delta}_r} \sim (p_c - p)^{-\hat{\gamma}_0'} |h|^{-\hat{\gamma}_0'/\hat{\Delta}'} (p_c - p)^{-\hat{\Delta}'(-\hat{\gamma}_0'/\hat{\Delta}')} = |h|^{-\hat{\gamma}_0'/\hat{\Delta}'}, \quad |h|^{-1/\hat{\delta}_i} \sim |h|^{-\hat{\gamma}_0'/\hat{\Delta}'}. \tag{3.37}$$

Therefore, by Lemma 10, $\hat{\delta}_r = \hat{\delta}_i = \hat{\Delta}'/\hat{\gamma}_0' = \hat{\delta}$. It is worth mentioning that these scaling relations are independent of the path of the limit $h \longrightarrow 0$.

Equations (2.18) and (2.24) imply that $m(p_c, h) \sim \lim_{p \longrightarrow p_c^+} h\hat{g}(p,h)$, for $0 < |h| \ll 1$. Therefore equations (3.7) and (3.34) imply that

$$|h|^{1/\delta_r} \sim m_r(p_c, h) = (p - p_c)^{\hat{\Delta} - \hat{\gamma}_0} \lim_{p \longrightarrow p_c^+} [\hat{x}_r \hat{\mathcal{G}}_{0,0}(\hat{x}) + |\hat{x}_r|^2 \hat{\mathcal{G}}_{0,1}(\hat{x})], \tag{3.38}$$
$$|h|^{1/\delta_i} \sim m_i(p_c, h) = (p - p_c)^{\hat{\Delta} - \hat{\gamma}_0} \lim_{p \longrightarrow p_c^+} [\hat{x}_i \hat{\mathcal{G}}_{0,0}(\hat{x})].$$

The analyticity of $g(p,h)$ and $\hat{g}(p,h)$ implies that they are bounded for all $p \in [0,1]$. Therefore, in order to cancel the diverging p dependent prefactors in equations (3.38), we must have $[\hat{x}_r \hat{\mathcal{G}}_{0,0}(\hat{x}) + |\hat{x}_r|^2 \hat{\mathcal{G}}_{0,1}(\hat{x})] \sim \hat{x}_i \hat{\mathcal{G}}_{0,0}(\hat{x}) \sim |x|^{(\hat{\Delta}-\hat{\gamma}_0)/\hat{\Delta}}$ as $\hat{x} \longrightarrow \infty$ ($p \longrightarrow p_c^+$ and $0 < h \ll 1$). Therefore equation (3.38) implies that $\delta_r = \delta_i = \hat{\Delta}/(\hat{\Delta} - \hat{\gamma}_0)$ *in general*, and for $1 \leq n_{\hat{\phi}} \leq n_\phi$, Lemmas 7 and 10 imply that $\delta_r = \delta_i = \hat{\Delta}/(\hat{\Delta} - \hat{\gamma}_0) = \Delta/(\Delta - \gamma) = \delta$. As before, these scaling relations are independent of the path of $h \longrightarrow 0$ □.

Lemma 13 *For* $1 \le n_{\hat\phi} \le n_\phi$*, the measure* $y\,d\phi(y)$ *has the symmetry property (*$\Delta = \Delta'$ *and* $\gamma = \gamma'$*) if and only if the measure* $d\hat\phi(y)$ *has the symmetry property (*$\hat\Delta = \hat\Delta'$ *and* $\hat\gamma_0 = \hat\gamma_0'$*). If either measure has this symmetry, then*

$$\mathbf{1})\quad s + t = \Delta\,. \quad \mathbf{2})\quad 1/\delta + 1/\hat\delta = 1. \quad \mathbf{3})\quad \Delta = \hat\Delta = \Delta' = \hat\Delta'. \quad \mathbf{4})\quad \gamma = \gamma' = \hat\gamma_0 = \hat\gamma_0'.$$

Proof: We have shown in Lemmas 7 and 8 that, for $1 \le n_{\hat\phi} \le n_\phi$, $\gamma = \hat\gamma_0$, $\Delta = \hat\Delta$, $\gamma' = \hat\gamma_0'$, and $\Delta' = \hat\Delta'$. Therefore, it is clear that, ($\Delta = \Delta'$ and $\gamma = \gamma'$) $\iff$ ($\hat\Delta = \hat\Delta'$ and $\hat\gamma_0 = \hat\gamma_0'$). Assume that either of the measures, $d\hat\phi(y)$ or $y\,d\phi(y)$, has this symmetry. Thus, $\Delta = \hat\Delta = \hat\Delta' = \Delta'$ and $\gamma = \hat\gamma_0 = \hat\gamma_0' = \gamma'$. By Lemma 9 we have $t = \Delta - \gamma$ and $s = \hat\gamma_0'$, and by Lemma 10 we have $\delta = \Delta/(\Delta - \gamma)$ and $\hat\delta = \hat\Delta'/\hat\gamma_0'$. Therefore,

$$\begin{aligned} &s + t = \hat\gamma_0' + \Delta - \gamma = \hat\gamma_0 + \Delta - \gamma = \Delta\,. \\ &\delta = \Delta/(\Delta - \gamma) = 1/(1 - \gamma/\Delta) = 1/(1 - \hat\gamma_0/\hat\Delta) = 1/(1 - \hat\gamma_0'/\hat\Delta') = 1/(1 - 1/\hat\delta) \quad \square. \end{aligned}$$

As mentioned above, the scaling relations $t_r = t_i = t$ and $s_r = s_i = s$ that we proved in Lemma 11 are fundamental identities, and serve as a consistency check of this mathematical framework. Another consistency check was given in Lemma 13, where we proved that $1/\delta + 1/\hat\delta = 1$. This is also a fundamental identity which follows from the relation in (2.24), $m(p, h) = h\,w(p, z(h))$, and the definition of these critical exponents in (3.7) and (3.8), for $0 < |h| \ll 1$: $h^{1/\delta} \sim m(p_c, h) = h\,w(p_c, z(h)) \sim h\,h^{-1/\hat\delta} \sim h^{1-1/\hat\delta}$. It follows that the relation in (2.24) provides a partial converse to the assumption underlying Lemma 13. Indeed as $1/\delta + 1/\hat\delta = 1$ in general, and for all $1 \le n_{\hat\phi} \le n_\phi$ we have $\delta = \Delta/(\Delta - \gamma) = \Delta/t$ and $\hat\delta = \hat\Delta'/\hat\gamma_0' = \hat\Delta'/s$, then $1 - \gamma/\Delta = 1/\delta = 1 - 1/\hat\delta = 1 - \hat\gamma_0'/\hat\Delta'$ implies that $t/\Delta + s/\hat\Delta' = 1$, and $\Delta = \hat\Delta' \iff \gamma = \hat\gamma_0'$. This concludes the proof of Theorem 5 $\square$.

3.3 Spectral Gaps and Critical Behavior of Transport

We now discuss the gaps θ_μ and θ_α in the spectral measures μ and α, respectively. As the operators Γ and Υ are projectors on the associated Hilbert spaces $\mathscr{H}_\times$ and $\mathscr{H}_\bullet$, respectively, their eigenvalues are confined to the set $\{0, 1\}$ [101]. The associated operators $M_j = \chi_j \Gamma \chi_j$ and $K_j = \chi_j \Upsilon \chi_j$, $j = 1, 2$ are positive definite compositions of projection operators, thus their eigenvalues are confined to the set $[0, 1]$ [65, 104]. While in general, the spectrum actually extends all the way to the spectral endpoints $\lambda = 0, 1$, it has been argued that the part close to $\lambda = 0, 1$ corresponds to very large, but very rare connected regions (Lifshitz phenomenon). It is believed that this phenomenon gives exponentially small contributions to the effective complex conductivity (resistivity), and does not affect power law behavior [60].

In [23] O. Bruno has proven the existence of spectral gaps in matrix/particle systems with polygonal inclusions, and studied how the gaps vanish as the inclusions touch (like $p \longrightarrow p_c$). In Figure 3.1 and Figure 3.2 we give graphical representations of the components α_{ik} of the (symmetric) tensor valued spectral measure $\boldsymbol{\alpha}$ for finite, square 2-d and 3-d RRN with independent and identically distributed (i.i.d.) x and y bonds (explained in more detail below). In the 2-d RRN, as $p \longrightarrow p_c = 0.5$ the gaps in the spectrum of the diagonal components α_{ii} of $\boldsymbol{\alpha}$ near $\lambda = 0, 1$ shrink to 0 *symmetrically.* In the 3-d RRN, as $p \longrightarrow p_c \approx 0.2488$ the spectral gap near $\lambda = 0$ shrinks to 0, and as $p \longrightarrow 1 - p_c \approx 0.7512$ the spectral gap near $\lambda = 1$ shrinks to 0. As p surpasses p_c and $1 - p_c$ the spectrum piles up at $\lambda = 0$ and $\lambda = 1$, respectively, forming delta function-like components in the measure. In Section 3.2.1 we showed that, for EMT, there are gaps in the spectral measures μ and α for $p \neq p_c, 1-p_c$. The gaps in μ and α about $\lambda = 1$ and $\lambda = 0$, respectively, collapse as $p \longrightarrow p_c$, and the gaps in μ and α about $\lambda = 0$ and $\lambda = 1$, respectively, collapse as $p \longrightarrow 1 - p_c$.

This is the behavior displayed in Lemma 2 and Corollary 1, which hold for general percolation models of stationary two-phase random media with $m(0) = m(p, 0)$ and $w(0) = w(p, 0)$. In this way the spectral measures μ and α truly are independent of the material contrast ratio, and are independent of how we define it. For example, we have focused on the contrast ratio $h = \sigma_1/\sigma_2$ and defined an insulator-conductor system by letting $\sigma_1 \longrightarrow 0$, resulting in critical behavior (the formation of a delta component in μ at $\lambda = 1$ with weight $m(p, 0)$) as p surpasses p_c, where $p = \langle\chi_2\rangle$ (see Lemma 2). We could have instead focused on $z = \sigma_2/\sigma_1$ and defined an insulator-conductor system by letting $\sigma_2 \longrightarrow 0$, resulting in critical behavior (the formation of a delta component in α at $\lambda = 1$ with weight $w(p, 0)$) as p surpasses $1 - p_c$ (see Corollary 1). Lemma 2 and Corollary 1 demonstrate, through spectral means, the equivalence of these two systems. Moreover these lemmas rigorously prove, for general percolation models of two-phase stationary random media in lattice and continuum settings, that the onset of critical behavior in transport is identified with the formation of delta components in μ and α at $\lambda = 0, 1$ *precisely* at $p = p_c$ and $p = 1 - p_c$.

For bond lattice systems with a finite number of bonds, N say, the differential equations in (2.3) become difference equations (Kirchoff's laws) [59]. Consequently, the operators M_j, $j = 1, 2$ are given [59, 64] by $n \times n$ matrices, say, and the spectral measure $\alpha_{ik}(d\lambda)$ of the

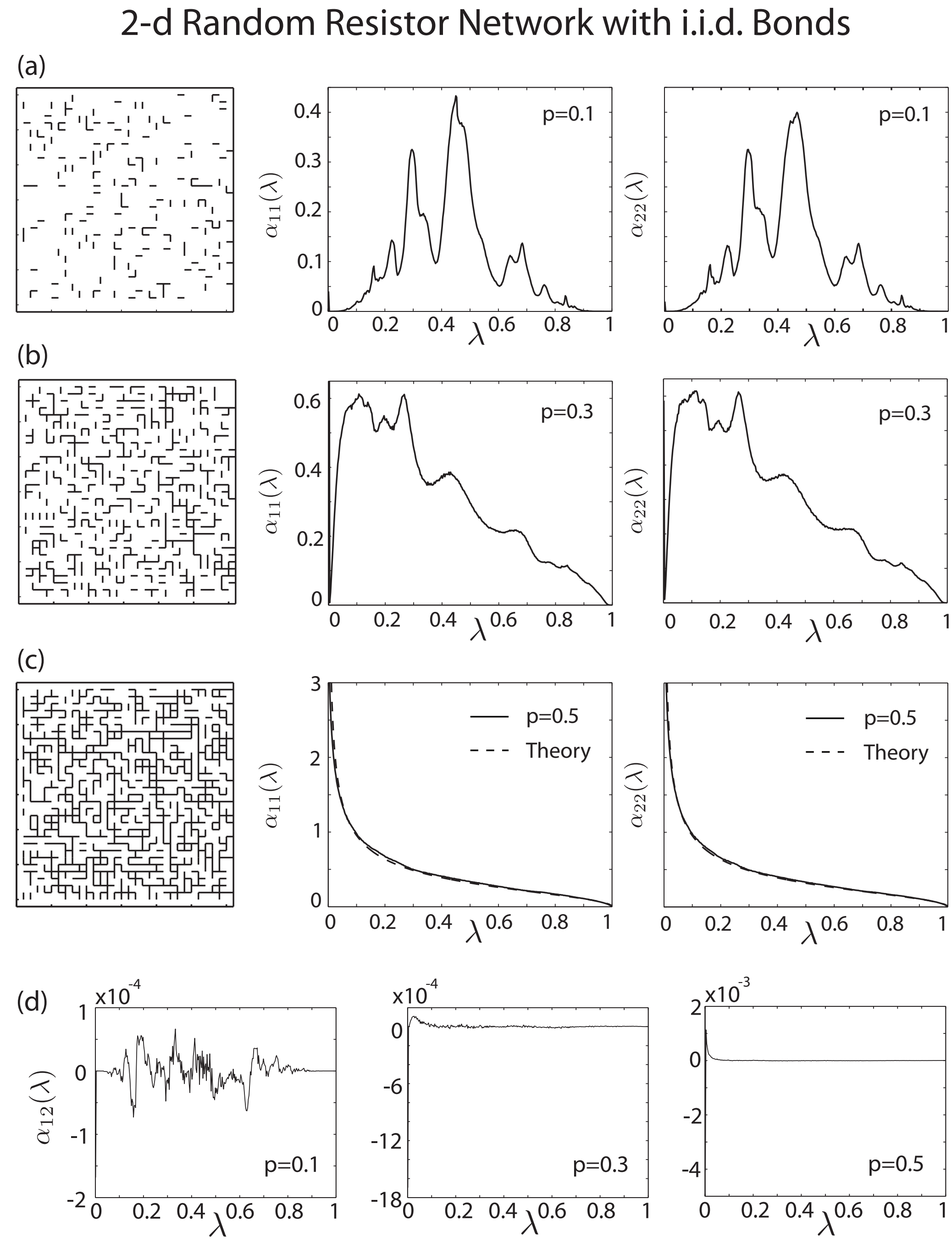

Figure 3.1. In the 2-d RRN (a)-(d), as the volume fraction p increases from top to bottom, the width of the gaps in the spectrum near $\lambda = 0, 1$ shrink to 0 *symmetrically* with increasing connectedness as $p \longrightarrow p_c = 1 - p_c = 0.5$. In (c) the effective medium theory prediction of the spectral measure, which coincides with the exact duality prediction, is also displayed. Consistent with the isotropy of the system, to numerical accuracy and finite size affects, the (positive) diagonal components $\alpha_{11}(\lambda)$ and $\alpha_{22}(\lambda)$ of the spectral functions in (a)–(c) are identical, and the (signed) off-diagonal component $\alpha_{12}(\lambda)$ in (d) is zero.

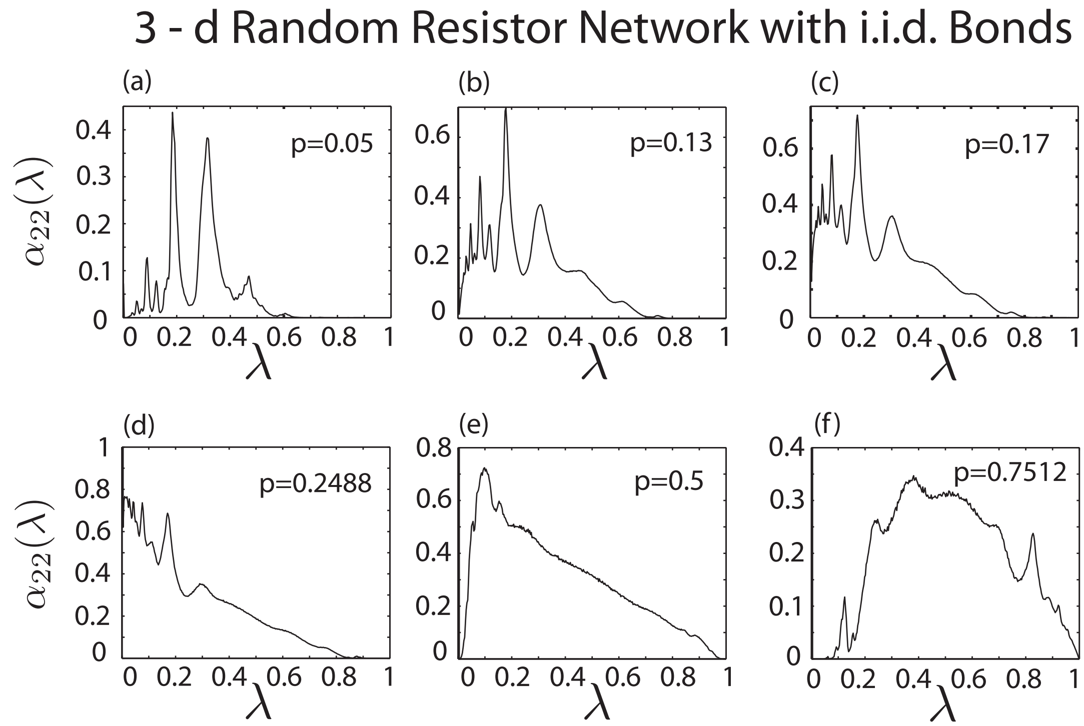

Figure 3.2. In the 3-d RRN (a)–(f), as the volume fraction p increases from let to right and top to bottom, the width of the gaps in the spectrum near $\lambda = 0, 1$ shrink to 0. As $p \longrightarrow p_c \approx 0.2488$ the width of the gap near $\lambda = 0$ shrinks to 0, and as $p \longrightarrow 1 - p_c \approx 0.7512$ the width of the gap near $\lambda = 1$ shrinks to 0. Similar to the 2-d RRN shown above, to numerical accuracy and finite size affects, the (positive) diagonal components $\alpha_{11}(\lambda)$, $\alpha_{22}(\lambda)$, and $\alpha_{33}(\lambda)$ of the spectral functions in (a)–(f) are identical, and the (signed) off-diagonal components $\alpha_{12}(\lambda)$ and $\alpha_{13}(\lambda)$ are zero, which is consistent with the isotropy of the RRN.

matrix M_2 is given by a sum of "Dirac δ functions,"

$$\alpha_{ik}(d\lambda) = \left[\sum_{j=1}^{n} m_j \delta_{\lambda_j}(d\lambda)\right] d\lambda = \alpha_{ik}(\lambda) d\lambda, \tag{3.39}$$

where $\delta_{\lambda_j}(d\lambda)$ is the delta measure concentrated at λ_j, $m_j = \langle \vec{e}_i^{\,T} [\vec{v}_j \vec{v}_j^{\,T}]\, \vec{e}_k \rangle$, $\vec{e}_k$ is a standard basis vector on the lattice, for some $k = 1, \ldots, d$, and λ_j and $\vec{v}_j$ are the eigenvalues and eigenvectors of M_2, respectively [64]. The associated Stieltjes transform of the measure in (2.13) is given by the sum $G(t) = \sum_{j=1}^{n} m_j/(t - \lambda_j)$, and $\alpha_{ik}(\lambda)$ in equation (3.39) is called "*the spectral function*," which is defined only pointwise on the set of eigenvalues $\{\lambda_j\}$.

In Figure 3.1 and Figure 3.2 we give graphical representations of the components α_{ik} of the tensor valued spectral measure $\boldsymbol{\alpha}$ for finite, square 2-d and 3-d RRN with i.i.d. x and y bonds. These figures display linearly connected peaks of histograms with bin sizes on the order of 10^{-2}. The apparent smoothness of these graphs in is due to the large number ($\sim 10^6$) of eigenvalues and eigenvectors calculated, and ensemble averaged. Consistent with the isotropy of these RRN, for each p, the diagonal components α_{kk} in Figures 3.1 and 3.2 are identical, positive measures of equal mass $1/d$, while the α_{ik}, $i \neq k$, are signed measures of zero mass, up to numerical accuracy and finite size effects. The α_{kk} do not have mass p, as the eigenvectors are normalized in the l_2 inner product, not that weighted by the characteristic function χ_2, like in the general theory. The mass of the measures have been scaled in these figures to have volume fraction p.

Figures 3.3-3.5 display graphical representations of α_{ik} for 2-d RRN that have i.i.d. x bonds with volume fraction p_x and i.i.d. y bonds with volume fraction p_y, with $p_x + p_y = p$. Consistent with the anisotropy of the RRN, the diagonal components are different and, consistent with the i.i.d. nature of the x and y bonds, the α_{ik} for $i \neq k$ are signed measures of zero mass, up to numerical accuracy and finite size effects. For $p_x = 0.5$ we recover the α_{ik} of Figure 3.1, and for $p_x > 0.5$ the measures $\alpha_{11}(\lambda)$ and $\alpha_{22}(\lambda)$ switch roles, which is consistent with the symmetry of the RRN.

We now provide an analytical proof for the existence of spectral gaps in α_{ik} about the spectral endpoints $\lambda = 0, 1$ for arbitrary, finite lattice systems. More specifically, for $p \ll 1$, $\inf\{\Sigma_\alpha\} > 0$ and $\sup\{\Sigma_\alpha\} < 1$. We focus on $M_2 = \chi_2 \Gamma \chi_2$ and α_{ik}, as our results extend to $M_1 = \chi_1 \Gamma \chi_1$ and μ_{ik} by symmetry. In this lattice setting, Γ is a real symmetric projection matrix which can be diagonalized: $\Gamma = QDQ^T$, where $D = \text{diag}(1,\ldots,1,0,\ldots,0)$ is a diagonal matrix of L ones and $n - L$ zeros, $0 < L < n$ when $n \gg 1$, and Q is a real orthogonal matrix with columns $\vec{q}_i$, $i = 1, \ldots, n$, which are the eigenvectors of Γ. More specifically,

$$\Gamma_{ij} = (\vec{q}_i \cdot \vec{q}_j)_L$$

where $(\vec{q}_i \cdot \vec{q}_j)_L = \sum_{l=1}^{L} (\vec{q}_i)_l (\vec{q}_j)_l$, and $(\vec{q}_i)_l$ is the l^{th} component of the vector $\vec{q}_i \in \mathbb{R}^n$. Here, we consider the nondegenerate case $L < n$.

In the matrix case, the action of χ_2 is given by that of a square diagonal matrix of zeros and ones [64]. The action of χ_2 in the matrix $\chi_2 \Gamma \chi_2$ introduces a row and column of zeros in the matrix Γ, corresponding to every diagonal entry of χ_2 with value 0. When there is only one σ_2 inclusion ($p = 1/N$) located at the j^{th} bond, χ_2 has all zero entries except at the j^{th} diagonal: $\chi_2 = \text{diag}(0, \cdots, 0, 1, 0, \cdots, 0) = \text{diag}(\vec{v}_j)$. Therefore, the only nontrivial eigenvalue is given by $\hat{\lambda}_0 = (\vec{q}_j \cdot \vec{q}_j)_L = \sum_{l=1}^{L} (\vec{q}_j)_l^2 = 1 - \sum_{l=L+1}^{n} (\vec{q}_j)_l^2$, with eigenvector $\vec{v}_j$

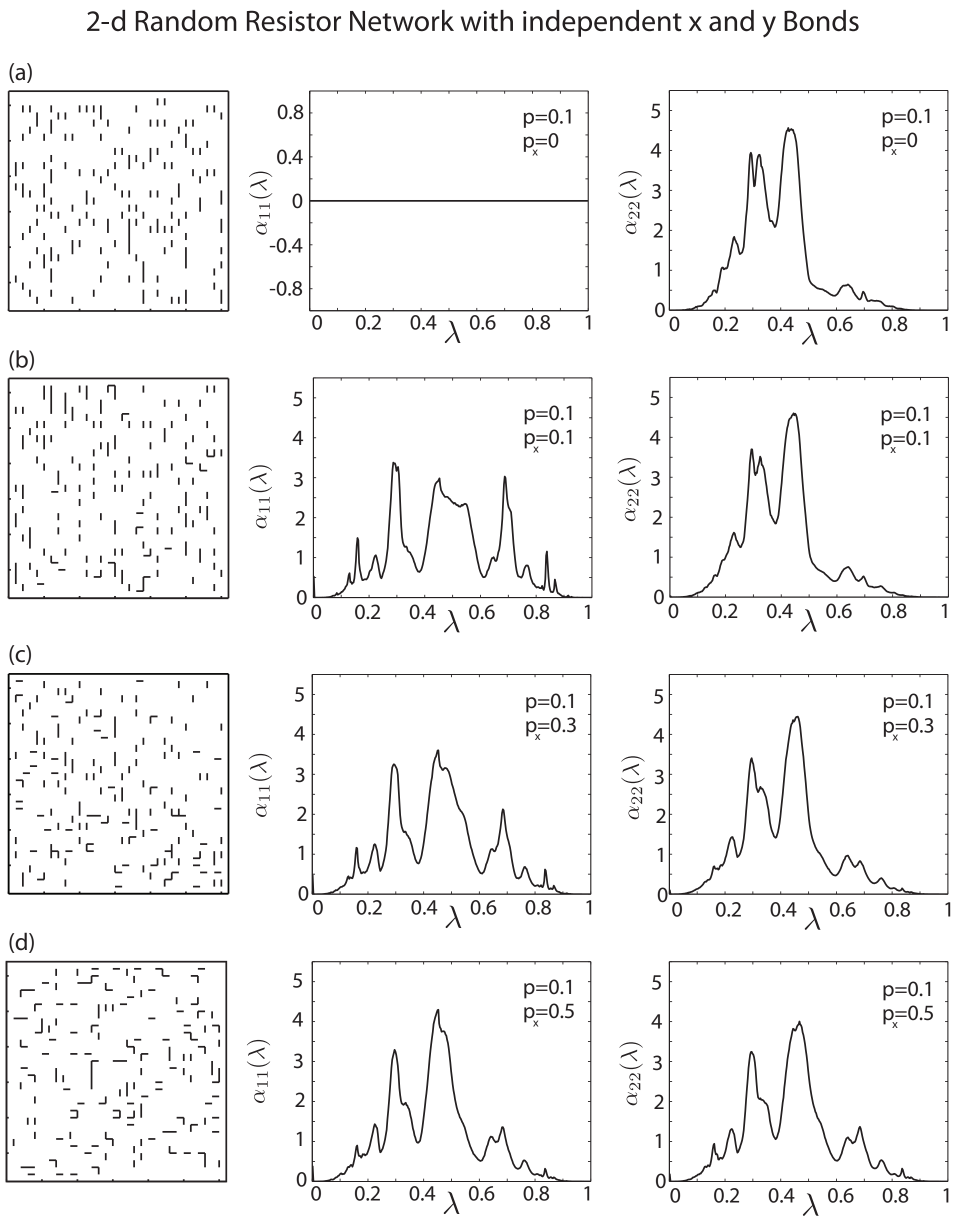

Figure 3.3. Random realizations of anisotropic 2-d square RRN with corresponding spectral functions $\alpha_{11}(\lambda)$ and $\alpha_{22}(\lambda)$ to the right. These RRN have i.i.d. x bonds with volume fraction p_x and i.i.d. y bonds with volume fraction p_y, with $p_x + p_y = p$. These RRN are disconnected for $p = 0.1$ and the spectral functions have gaps in the spectra near $\lambda = 0, 1$. For $p_x = 0$ the x component of the spectral function $\alpha_{11}(\lambda)$ is identically zero.

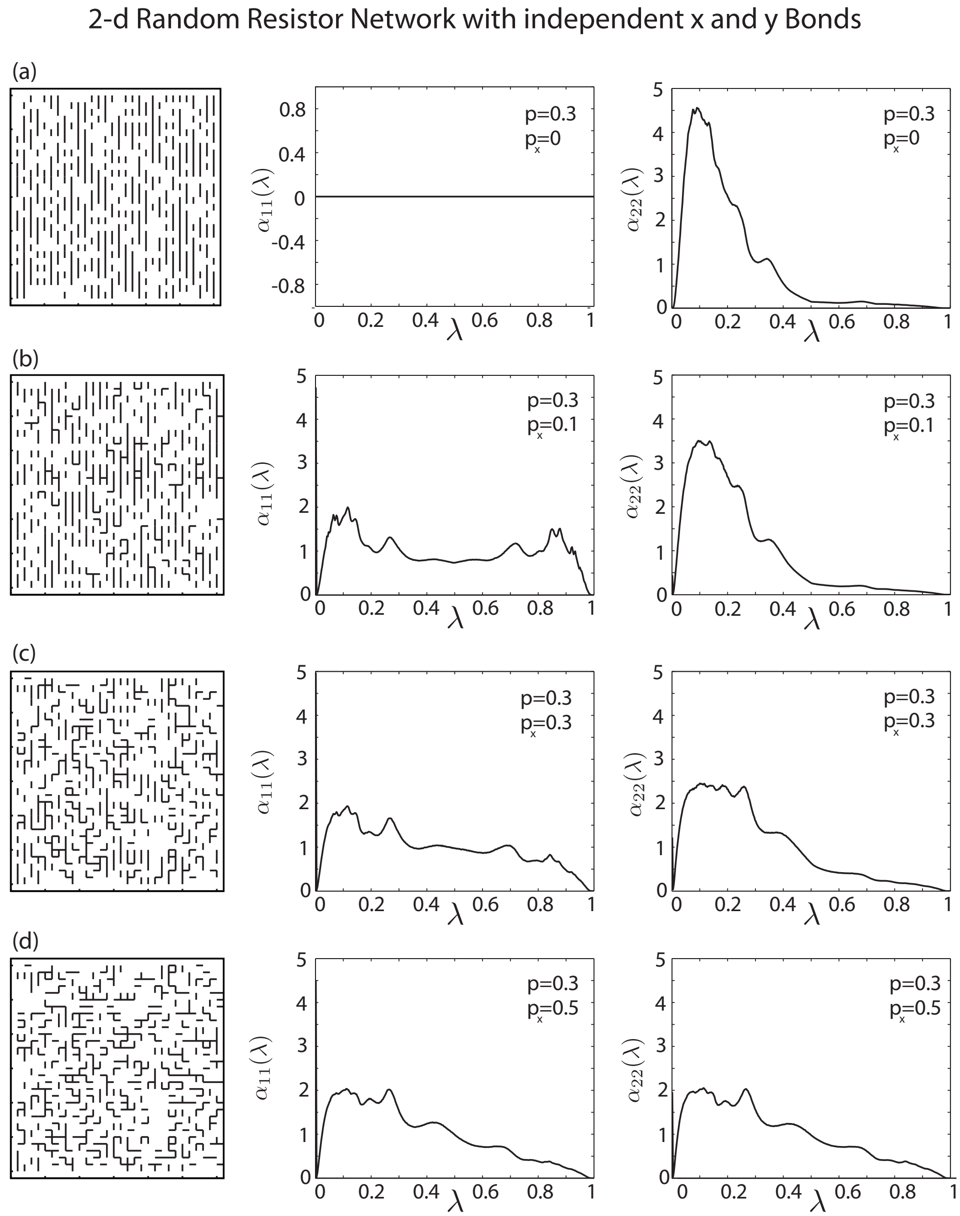

Figure 3.4. Random realizations of anisotropic 2-d square RRN with corresponding spectral functions $\alpha_{11}(\lambda)$ and $\alpha_{22}(\lambda)$ to the right. These RRN have i.i.d. x bonds with volume fraction p_x and i.i.d. y bonds with volume fraction p_y, with $p_x + p_y = p$. These RRN are disconnected for $p = 0.3$ and the spectral functions have gaps in the spectra near $\lambda = 0, 1$. However, these gaps are smaller than those displayed in Figure 3.3 where $p = 0.1$. For $p_x = 0$ the x component of the spectral function $\alpha_{11}(\lambda)$ is identically zero.

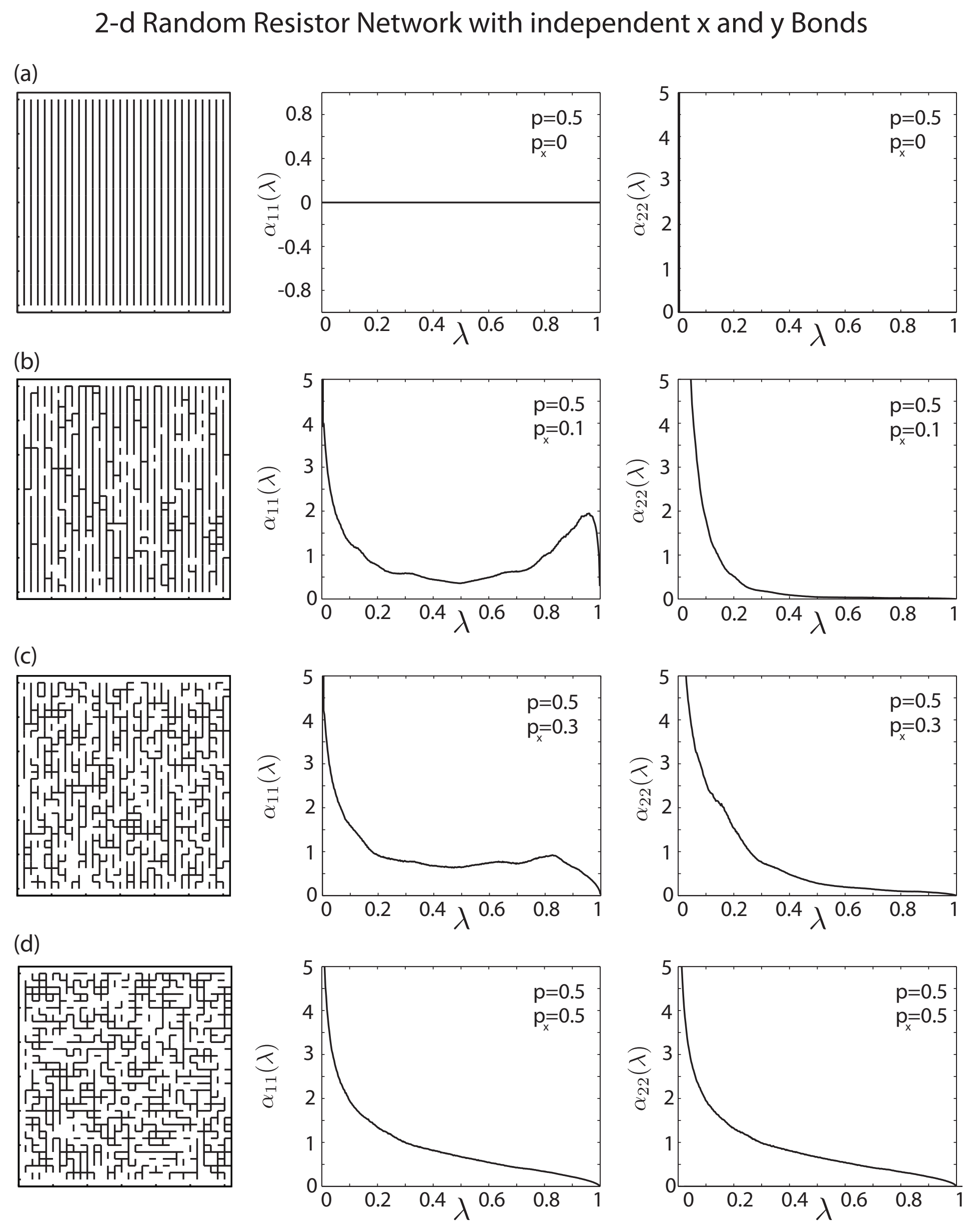

Figure 3.5. Random realizations of anisotropic 2-d square RRN with corresponding spectral functions $\alpha_{11}(\lambda)$ and $\alpha_{22}(\lambda)$ to the right. These RRN have i.i.d. x bonds with volume fraction p_x and i.i.d. y bonds with volume fraction p_y, with $p_x + p_y = p$. These RRN are connected in the x and y directions for $p = 0.5$ and the gaps in spectra near $\lambda = 0, 1$, displayed in Figures 3.3 and 3.4, have shrunk to 0. For $p_x = 0$ the x component of the spectral function $\alpha_{11}(\lambda)$ is identically zero, while the y component of the spectral function $\alpha_{22}(\lambda)$ is a delta function concentrated at $\lambda = 0$.

and weight $m_0 = \vec{e}_i^{\,T}\vec{v}_j\vec{v}_j^{\,T}\vec{e}_k$. This implies that there is a gap at $\lambda = 0$, $\theta_0 = \sum_{l=1}^{L}(\vec{q}_j)_l^2 > 0$, and a gap at $\lambda = 1$, $\theta_1 = \sum_{l=L+1}^{n}(\vec{q}_j)_l^2 > 0$. It is clear that these bounds hold for all $\omega \in \Omega$ such that $p = 1/N$ when $L < n$. We have already mentioned that the eigenvalues of M_2 are restricted to the set $\{0, 1\}$ when $p = 1$ ($\chi_2 \equiv I_n$). Therefore, there exists $0 < p_0 < 1$ such that, for all $p \geq p_0$, there exists a $\omega \in \Omega$ such that $\theta_0(\omega) = 0$ and/or $\theta_1(\omega) = 0$. This concludes our proof $\square$.

3.4 Concluding Remarks

We have constructed a mathematical framework which unifies the critical theory of transport for binary composite media, in continuum and lattice settings. We have focused on critical transitions exhibited by the effective complex conductivity $\sigma^* = \sigma_2 m(h) = \sigma_1 w(z)$, as the symmetries underlying this framework extend our results to that regarding the effective complex resistivity $\rho^* = \tilde{m}(h)/\sigma_1 = \tilde{w}(z)/\sigma_2$. We have shown in Section 2.3 that critical transitions in transport properties are, in general, characterized by the formation of delta components in the underlying spectral measures at the spectral endpoints. Moreover, for percolation models, we have shown that the onset of the critical transition (the formation of these delta components) occurs *precisely* at the percolation threshold p_c and $1 - p_c$.

The mathematical transport properties of such systems, displayed in Section 2.1 and Section 2.2, hold for general two-component stationary random media in lattice and continuum settings [65]. While the critical exponent scaling relations and the various transport properties, displayed in Lemmas 4–13, hold for percolation models of the composites class $\mathcal{B}_{n_\phi}$ and $\mathcal{B}_{n_{\hat{\phi}}}$ for $p > p_c$ and $p < p_c$, respectively (see Definition 1). Under the condition that $n_\phi, n_{\hat{\phi}} \geq 1$, i.e., that $m(p, 0)$ and $\chi(p, 0)$ in (3.7) exist for $p > p_c$, and $w(p, z(0))$ and $\hat{\chi}(p, z(0))$ in (3.8) exist for $p < p_c$, we linked the two sets of critical exponents in (3.7) and (3.8). We showed that, for percolation models like EMT where $n_\phi, n_{\hat{\phi}} = \infty$, they are all determined by only three critical exponents, and are determined by only two critical exponents under the symmetry condition of Lemma 13. This type of critical behavior has been studied before for the lattice [13, 37, 52], and alternate methods have shown that $\Delta = s + t$, $\delta = (s + t)/t$, and $\gamma = s$. These are precisely the relations that we have shown to hold for lattice and continuum percolation models of this class, under these conditions. The EMT percolation model satisfies these conditions, however, there is no apparent mathematical necessity for these conditions to hold, in general. Although they lead to the well known two-dimensional duality relation $s = t$ for the lattice [13, 37, 52].

Numerical and analytical work on the sequence of critical exponents $\tilde{\psi}(q)$ for the moments of the current distribution in RRN, e.g., [17, 45, 116], has shown that this sequence exhibits nonlinear dependence in q, or multifractal behavior. We proved in Lemma 9 that the exponents γ_n have linear dependence in n for all $1 \leq n \leq n_{\hat{\phi}} \leq n_\phi$. This is consistent with the absence of multifractal behavior for percolation models of class $\mathcal{B}_\infty$, such as EMT. However for percolation models with $n_{\hat{\phi}}, n_\phi < \infty$, multifractal behavior is not ruled out by Lemma 9. It is interesting, though, that the $\tilde{\psi}(q)$ satisfy Baker's inequalities (3.13) [17].

As in EMT, our general scaling relations involving $|h|$ are independent of the limiting path as $h \longrightarrow 0$. This represents an alternative to the results of other workers [13, 37, 52] who have used heuristic scaling forms as a starting point. For our critical theory the starting point is equation (2.13), which displays *exact* formulas for infinite systems [60].

CHAPTER 4

ORTHOGONAL POLYNOMIALS

The theory of orthogonal polynomials is intimately entwined within the spectral theory of bounded self-adjoint linear operators with simple spectrum [41, 118]. In this chapter we explore the connection between orthogonal polynomials and the theory of two-phase random media. We introduce numerator and denominator polynomials in Section 4.1 by reviewing their role in Padé approximants of $F(s;\mu)$, the Stieltjes transform of the measure μ introduced in Section 2.1. In Section 4.2 we review the theory of denominator polynomials and provide a mapping structure, associated with two-phase stationary random media, between various measure and operator spaces. In Section 4.3 we give the key results of this chapter. There, we introduce generalized numerator polynomials $P^{[i]}$, $i=0,1,\ldots$, and we demonstrate the physical significance of the Stieltjes transforms $F^{[i]}(s;\mu^{[i]})$ of the orthogonality measures $\mu^{[i]}$ associated with these families of polynomials. More specifically, for $i=0,1,2$, the $F^{[i]}(s;\mu^{[i]})$ are completely determined by components of the energy decomposition given in Theorem 4. Moreover, we derive a novel, closed form solution for the moments $\mu_j^{[i]}$ of the measures $\mu^{[i]}$ in terms of the moments μ_j of μ, for all $i\in\mathbb{N}$, $j=0,1,\ldots$. This novel solution for the $\mu_j^{[i]}$ uniquely determines these measures, as moment problem is determined for the measures $\mu^{[i]}$ [35, 114].

4.1 Padé Approximants

In Section 2.1 we derived integral representations for the effective complex conductivity and resistivity tensors $\boldsymbol{\sigma}^*$ and $\boldsymbol{\rho}^*$, respectively, which involve Stieltjes transforms of the tensor valued measures $\boldsymbol{\mu}$, $\boldsymbol{\alpha}$, $\boldsymbol{\eta}$, and $\boldsymbol{\kappa}$. These are spectral measures associated with the bounded linear operators $M_j=\chi_j\Gamma\chi_j$ and $K_j=\chi_j\Upsilon\chi_j$, $j=1,2$, which are self-adjoint on $L^2(\Omega,P)$. Following Van Assche [125], in this section we review Padé approximants of the Stieltjes transform of a measure. We will limit our attention to the Stieltjes transform of the measure $\boldsymbol{\mu}$ associated with the operator $M=M_1=\chi_1\Gamma\chi_1$, as results associated with the other operators and measures follow by symmetry. For simplicity, we focus on a diagonal component $\mu = \mu_{kk}$ of the measure and its Stieltjes transform $F(s;\mu) = F_{kk}(s;\mu)$ associated with $\sigma^* = \sigma^*_{kk}$, for some $k = 1, \ldots, d$.

Geometric information of a two-phase random medium is incorporated into the measure μ through its moments μ_j, $j = 0, 1, \ldots$, which depend on the correlation functions of the medium [65]. This can be seen by expanding σ^* around a homogeneous medium $\sigma_1 = \sigma_2$ ($s = \infty$), or equivalently expanding the *integral* for $F(s;\mu)$ in (2.13) in powers of $1/s$ for $|s| > 1$, yielding [59]

$$F(s) = \sum_{j=0}^{\infty} \frac{\mu_j}{s^{j+1}} \,. \tag{4.1}$$

Performing the same s expansion for $F(s;\mu) = \langle \chi_1 (s - \Gamma\chi_1)^{-1} \vec{e}_k \cdot \vec{e}_k \rangle$ yields [59, 65]

$$\mu_j = \int_0^1 \lambda^j d\mu(\lambda) = \int_\Omega P(d\omega)[\chi_1(\Gamma\chi_1)^j \vec{e}_k] \cdot \vec{e}_k, \quad j = 0, 1, \ldots \tag{4.2}$$

Clearly, in general we have

$$\mu_0 = \langle \chi_1 \rangle = p_1, \tag{4.3}$$

where p_1 is the volume fraction of the σ_1 phase, with $p_2 = 1 - p_1$, where $\langle\cdot\rangle$ denotes ensemble average over Ω. When the medium is isotropic it can be shown that [24, 65]

$$\mu_1 = \frac{p_1 p_2}{d} \,. \tag{4.4}$$

Equation (4.2) shows that μ_n depends on the $(n+1)$-point correlation function of the medium under consideration [59, 65]. In principle, if all the n-point correlation functions are known then the spectral measure may be uniquely determined and $F(s)$, hence σ^*, is exactly known [114]. This shows the usefulness of the representation (4.1): by analytic continuation, local information at $s = \infty$ yields global information [87] for $s \in \mathbb{C}\backslash\Sigma_\mu$, where the support of μ is denoted $\Sigma_\mu = \mathrm{supp}(\mu) \subseteq [0, 1]$.

In practice, complete information regarding the composite is unavailable and one often resorts to approximations of $F(s) = F(s;\mu)$. Taylor polynomials are clearly not a good class of functions to approximate $F(s)$ as they do not capture the singularities of this function. Rational functions of polynomials are the simplest approximating functions with singularities [125]. The $[m, n]$ Padé approximant of $F(s)$ is the rational function of *monic* polynomials Q_m/P_n, with Q_m of degree $\leq m$ and P_n of degree $\leq n$. For the Padé approximation of $F(s)$ near infinity one takes $m = n-1$, which leads to the following interpolation condition [125]:

$$P_n(s)F(s) - Q_{n-1}(s) = O(s^{-n-1}), \quad |s| \longrightarrow \infty. \tag{4.5}$$

The *denominator polynomials* P_n are orthogonal with respect to the measure μ [125],

$$\int_0^1 \lambda^k P_n(\lambda)d\mu(\lambda) = 0, \quad k = 0, 1, \ldots, n-1, \tag{4.6}$$

which we will normalize $\tilde{p}_n = P_n/\|P_n\|_\mu$, where $\|\cdot\|_\mu$ is the $L^2(\mu)$ norm. The corresponding numerator polynomials, $\tilde{q}_{n-1} = Q_{n-1}/\|P_n\|_\mu$ are given by [125]

$$\tilde{q}_{n-1}(s) = \int_0^1 \frac{\tilde{p}_n(s) - \tilde{p}_n(\lambda)}{s-\lambda} d\mu(\lambda), \tag{4.7}$$

and the error may be written as [125]

$$F(s) - \frac{\tilde{q}_{n-1}(s)}{\tilde{p}_n(s)} = \frac{1}{\tilde{p}_n^2(s)} \int_0^1 \frac{\tilde{p}_n^2(\lambda)}{s-\lambda} d\mu(\lambda). \tag{4.8}$$

In order to illuminate some important properties of orthogonal polynomials, we sketch a convergence analysis of (4.8) for s in a compact set $K \subset \mathbb{C}\backslash\Sigma_\mu$.

The integral on the right hand side of (4.8) may be bounded independent of n [125], so the convergence of the Padé approximants is completely determined by the asymptotic behavior of $\tilde{p}_n$ as $n \longrightarrow \infty$. By orthogonality, the zeros of $\tilde{p}_n$ are simple and contained in $(0,1)$ [41]. Denote them by $0 < \lambda_{1,n} < \lambda_{2,n} < \cdots < \lambda_{n,n} < 1$, and denote the leading coefficient of $\tilde{p}_n$ by $\gamma_n = \|P_n\|_\mu^{-1}$, so that

$$\tilde{p}_n(s) = \gamma_n \prod_{j=1}^n (s - \lambda_{j,n}). \tag{4.9}$$

The asymptotic behavior of $\tilde{p}_n$ is thus determined by that of γ_n and the asymptotic distribution of the zeros $\{\lambda_{j,n}\}$.

First, consider the normalized counting measure of the zeros

$$\nu_n(d\lambda) = \frac{1}{n} \sum_{j=1}^n \delta_{\lambda_{j,n}}(d\lambda), \tag{4.10}$$

which defines a sequence of probability measures supported on the interval $(0,1)$, and describes the distribution of the zeros of $\tilde{p}_n$. Helly's selection principle tells us [41, 125] that there is a subsequence $\{n_k\}$ that converges weakly to a probability measure ν supported on the interval $[0,1]$. For the monic polynomial P_n we have

$$\frac{1}{n}\log|P_n(s)| = \frac{1}{n}\sum_{j=1}^{n}\log|s-\lambda_{j,n}| = \int_0^1 \log|s-\lambda_{j,n}|d\nu_n(\lambda), \tag{4.11}$$

so that, by weak convergence for $s \in K$,

$$\lim_{j\longrightarrow\infty}|P_{n_j}(s)|^{1/n_j} = \exp\left(U_\nu(s;\nu)\right), \quad U_\nu(s;m) = \int_{\Sigma_\nu}\log|s-\lambda|dm(\lambda), \tag{4.12}$$

where $\Sigma_\mu \subseteq \Sigma_\nu \subseteq [0,1]$ and we use $\nu_n(d\lambda)$ and $d\nu_n(\lambda)$ interchangeably.

Second, just as the Chebyshev polynomials minimize the $L^\infty([-1,1])$ norm, the P_n minimize the $L^2(\mu)$ norm [41]. Indeed, the leading coefficient $\gamma_n = \|P_n\|_\mu^{-1}$ solves the minimization problem:

$$\gamma_n^{-2} = \inf\nolimits_{\pi_n(\lambda)=\lambda^n+\cdots}(\|\pi_n\|_\mu^2), \tag{4.13}$$

so that the minimum is attained at the monic orthogonal polynomial P_n [125]. This extremal problem for γ_n may be used to show that the asymptotic behavior of $\gamma_n^{1/n}$ and the asymptotic distribution ν of the zeros are described by an equilibrium problem for logarithmic potentials [125].

More specifically, if μ is sufficiently regular [108, 115] and if we denote by $\mathcal{M}_1(\Sigma_\nu)$ the set of probability measures supported on Σ_ν, then the measure ν is the unique minimizer (the equilibrium measure for Σ_ν) of the logarithmic functional

$$\mathcal{E}_\nu[\nu] = \inf_{m\in\mathcal{M}_1(\Sigma_\nu)}\mathcal{E}_\nu[m], \quad \mathcal{E}_\nu[m] = -\int_{\Sigma_\nu}U_\nu(\lambda;m)dm(\lambda), \tag{4.14}$$

were $U_\nu(\lambda;m)$ is defined in equation (4.12). This standard variational problem of potential theory, which has the electrostatic interpretation of ν being the equilibrium distribution of a distribution m of positive charges on a conductor Σ_ν, is equivalent [108] to the relations:

$$\begin{aligned} -2U_\nu(\lambda;\nu) &= -l_\nu, \quad \lambda\in\Sigma_\nu, \\ -2U_\nu(\lambda;\nu) &\geq -l_\nu, \quad \lambda\in\mathbb{R}\backslash\Sigma_\nu. \end{aligned} \tag{4.15}$$

Formulas (4.15) are the Euler–Lagrange equations for (4.14), the quantity $\exp(l_\nu/2)$ is the logarithmic capacity of Σ_ν, and $-l_\nu/2$ is known as the Robin constant [97]. It can be shown, on the potential curves

$$C_r = \left\{s\in\mathbb{C}\backslash\Sigma_\nu \mid \left(\lim_{n\longrightarrow\infty}\gamma_n^{1/n}\right)\exp(U_\nu(s;\nu)) = r\right\}, \tag{4.16}$$

with $r > 1$, that

$$\lim_{n\longrightarrow\infty}\left|F(s)-\frac{\tilde{q}_{n-1}(s)}{\tilde{p}_n(s)}\right|^{1/n}=\frac{1}{r^2}, \tag{4.17}$$

demonstrating that the convergence is exponential [125].

A Padé approximation of $F(s)$ thus gives an idea of the singularities $\{\lambda_j\}$ of $F(s)$. The singularities of the Padé approximant are the roots of the polynomial P_n. From equation (2.13) we see that $F(s)$ is a linear mapping of $\mathscr{B}_0$, the set of positive finite Borel measures on $[0,1]$, to the complex plane. Let

$$\mathscr{B}(\mu_1,\mu_2,\ldots,\mu_n)=\left\{\mu\in\mathscr{B}_0 \mid \ \mu_j=\int_0^1\lambda^j d\mu(\lambda),\ j=1,2,\ldots,n\right\}. \tag{4.18}$$

The set of measures $\mathscr{B}(\mu_1,\mu_2,\ldots,\mu_n)$ is a compact, convex subset of $\mathscr{B}_0$ with the topology of weak convergence [65]. One can show that the measure μ is a weak limit of convex combinations of n-point measures, i.e., measures of the form [65]

$$\mu(d\lambda)=\sum_{j=1}^n m_j\delta_{\lambda_j}(d\lambda), \tag{4.19}$$

where $d\mu(\lambda)$ and $\mu(d\lambda)$ are used interchangeably and

$$m_j\geq 0,\ 0\leq\lambda_1<\lambda_2<\cdots<\lambda_n<1,\ \mu_i=\sum_{j=1}^n m_j\lambda_j^i,\ i=0,1,\ldots,n-1. \tag{4.20}$$

In finite lattice systems, the random operator $M_n=M=\chi_1\Gamma\chi_1$ is a sparse *random matrix* [58] of size n, say. In this case, the discrete measure μ is given exactly by equation (4.19), and $F(s)$ may be written [58]

$$F(s)=\sum_{j=1}^n\frac{m_j}{s-\lambda_j},\quad m_j=\langle\vec{e}_k^{\,T}E_{\lambda_j}\vec{e}_k\rangle,\quad E_{\lambda_j}=\vec{v}_j\vec{v}_j^{\,T},\quad M_n\vec{v}_j=\lambda_j\vec{v}_j, \tag{4.21}$$

where $\vec{v}_i^{\,T}\vec{v}_j=\delta_{i,j}$, $\vec{e}_k$ is a standard basis vector on the lattice, and $\langle\cdot\rangle$ denotes ensemble average. The family of projection operators E_λ is called the *resolution of the identity*, $\sum_{j=1}^n E_{\lambda_j}=I_n$, where I_n is the identity operator on $\mathbb{R}^n$. In this finite lattice setting the Padé approximants of $F(s)$ are exact:

$$F(s)=p_1\frac{Q_{n-1}(s)}{P_n(s)},\quad P_n(s)=\prod_{j=1}^n(s-\lambda_j),\quad Q_{n-1}(s)=p_1^{-1}\sum_{j=1}^n m_j\prod_{j\neq l=1}^n(s-\lambda_l), \tag{4.22}$$

where the normalization by the volume fraction, $p_1=\sum_{j=1}^n m_j=\int_0^1 d\mu(\lambda)$, makes $Q_{n-1}(s)$ a monic polynomial. In this case, the singularities of $F(s)$ (the eigenvalues of M_n) are precisely the zeros of $P_n(s)$. By multiplying $F(s)$ in (4.21) by $s-\lambda_i$ and letting $s \longrightarrow \lambda_i$ one can easily see from (4.22) that the weights (Christoffel numbers) $\{m_j\}_{j=1}^n$ are given by

$$m_j = p_1 \frac{Q_{n-1}(\lambda_j)}{P_n'(\lambda_j)}, \tag{4.23}$$

where the prime denotes differentiation in the variable s.

4.2 Denominator Polynomials

In this section, from the spectral parameters $\{m_j, \lambda_j\}$ of the matrix M_n, we construct the Jacobi matrix J_n which determines the denominator polynomials P_j, $j = 0, \dots, n-1$. We also provide a mapping structure, associated with two-phase stationary random media, between various measure and operator spaces. In the infinite operator case, the spectral theorem [101] generates a mapping φ_μ from M to a spectral measure μ: $M \overset{\varphi_\mu}{\mapsto} \mu$. We discuss another (one-to-one) map $\mu \overset{\psi}{\mapsto} J$ from μ to a bounded Jacobi matrix J of infinite order which acts on l_2. The construction of ψ turns out to be the classical problem of generating a family of polynomials orthogonal to μ. This problem demonstrates that the inverse map $\varphi = \psi^{-1}$ is the spectral map $J \overset{\varphi}{\mapsto} \tau$ from the operator J to a positive measure τ, and it extends to a unitary map $l_2 \overset{U}{\mapsto} L^2(\mu)$. In the matrix case the map ψ becomes a bijection.

It is important to note that the measure μ defined by (4.19)–(4.21), for $n < \infty$, can only generate polynomials orthogonal to μ up to order $n-1$. This can be easily seen [41] as the polynomial $\pi(s) = \prod_{j=1}^n (s-\lambda_j) = 0$ in $L^2(\mu)$ so that $\{1, s, s^2, \dots, s^{n-1}\}$ spans $\{1, s, s^2, \dots\}$ in $L^2(\mu)$. Conversely [41] observe that for some i, the set of polynomials $\{1, s, s^2, \dots, s^i\}$ is linearly dependent in $L^2(\mu)$ only if $i < n$. Indeed, if this set is dependent for some $i \geq n$, then there exists a polynomial $\pi(s) = \sum_{j=1}^i c_j s^j$, with not all $c_j = 0$, such that $\pi(s) = 0$ in $L^2(\mu)$. But $\pi(s)$ has at most $i \geq n$ real zeros, a contradiction, hence $i < n$.

The Gram–Schmidt procedure can be carried out on the set $\{1, s, s^2, \dots\}$ as long as the $L^2(\mu)$ norm $\|\cdot\|_\mu$ is strictly positive definite. If we denote by $\mathbb{P}$ the set of real polynomials on $[0,1]$ and $\mathbb{P}_i \subset \mathbb{P}$ the space of polynomials of degree $\leq i$, then the above argument shows that $\|\cdot\|_\mu$ is strictly positive definite on $\mathbb{P}_i$ only if $i < n$. Moreover, the $L^2(\mu)$ norm is strictly positive definite on $\mathbb{P}_i$ if and only if the Hankel determinants D_j are strictly positive $D_j > 0$, for $j = 1, 2, \dots, i$, where

$$D_j = \begin{vmatrix} \mu_0 & \mu_1 & \mu_2 & \cdots & \mu_j \\ \mu_1 & \mu_2 & \mu_3 & \cdots & \mu_{j+1} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ \mu_j & \mu_{j+1} & \mu_{j+2} & \cdots & \mu_{2j} \end{vmatrix} \tag{4.24}$$

and the $\{\mu_j\}$ are the moments of the measure μ defined in (4.2). This can be easily seen [57] by writing $\pi_j(\lambda) = \sum_{j=1}^{i} c_j \lambda^j$ so that

$$\|\pi_j\|_\mu^2 = \int_0^1 d\mu(\lambda) \sum_{j,l=0}^{i} c_j c_l \lambda^{j+l} = \sum_{j,l=0}^{i} c_j c_l \mu_{j+l}. \tag{4.25}$$

Therefore, the norm $\|\cdot\|_\mu$ is strictly positive definite if and only if the Hankel matrix $[\mu_{j+l}]_{j,l=0,1,2,\cdots,i}$ is strictly positive definite if and only if $D_j > 0$ for $j = 1, 2, \ldots, i$.

In the case of an infinite two-phase random medium ($n = \infty$), an infinite sequence of orthogonal polynomials may be generated. The polynomials $\tilde{p}_i = P_i/\|P_i\|_\mu$ orthonormal in $L^2(\mu)$,

$$\langle \tilde{p}_i, \tilde{p}_j \rangle_\mu = \int_{\Sigma_\mu} \tilde{p}_i(\lambda) \tilde{p}_j(\lambda) d\mu(\lambda) = \delta_{i,j}, \tag{4.26}$$

are given by [41, 119]

$$\tilde{p}_i(s) = (D_{i-1} D_i)^{-1/2} \begin{vmatrix} \mu_0 & \mu_1 & \mu_2 & \cdots & \mu_i \\ \mu_1 & \mu_2 & \mu_3 & \cdots & \mu_{i+1} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ \mu_{i-1} & \mu_i & \mu_{i+1} & \cdots & \mu_{2i-1} \\ 1 & s & s^2 & \cdots & s^i \end{vmatrix}, \tag{4.27}$$

where D_i is defined in equation (4.24). The zeros of $\tilde{p}_{i-1}$ interlace those of $\tilde{p}_i$ [73], and $\tilde{p}_n'(s)/\tilde{p}_n(s) = \sum_{j=1}^{n} (s - \lambda_{j,n})^{-1}$ [124].

The polynomials $\tilde{p}_i$ satisfy the following three term recursion relation [57]

$$\begin{gathered} \sqrt{\beta_{j+1}} \tilde{p}_{j+1}(s) = (s - \alpha_j) \tilde{p}_j(s) - \sqrt{\beta_j} \tilde{p}_{j-1}(s), \quad j = 0, 1, 2, \ldots \\ \tilde{p}_{-1}(s) = 0, \quad \tilde{p}_0(s) = 1/\sqrt{\beta_0}, \end{gathered} \tag{4.28}$$

where [41, 57]

$$\begin{aligned} \alpha_j &= \frac{\langle \lambda P_j, P_j \rangle_\mu}{\langle P_j, P_j \rangle_\mu} = \langle \lambda \tilde{p}_j, \tilde{p}_j \rangle_\mu, \quad j = 0, 1, 2, \ldots \\ \beta_j &= \frac{\langle P_j, P_j \rangle_\mu}{\langle P_{j-1}, P_{j-1} \rangle_\mu} = \langle \lambda \tilde{p}_{j-1}, \tilde{p}_j \rangle_\mu^2, \quad j = 1, 2, \ldots, \end{aligned} \tag{4.29}$$

and $\beta_0 = \langle P_0, P_0 \rangle_\mu = \int_0^1 d\mu(\lambda) = p_1$, hence $\|P_n\|_\mu^2 = \beta_n \beta_{n-1} \cdots \beta_1 \beta_0$. Using the recursion coefficients $\{\alpha_j, \beta_j\}$, the numerator polynomial Q_{n-1} in (4.23) can be replaced, giving the Christoffel numbers m_j in terms of the $\tilde{p}_j$ and their derivatives [124]

$$m_j = p_1 \frac{Q_{n-1}(\lambda_{j,n})}{P_n'(\lambda_{j,n})} = -(\sqrt{\beta_{n+2}} \tilde{p}_{n+1}'(\lambda_{j,n}) \tilde{p}_{n+2}(\lambda_{j,n}))^{-1} = (\sqrt{\beta_{n+1}} \tilde{p}_{n+1}'(\lambda_{j,n}) \tilde{p}_n(\lambda_{j,n}))^{-1} \tag{4.30}$$

Denote the essentially self-adjoint Jacobi operator $J(\mu)$ [41, 57, 118] by

$$J(\mu) = \begin{bmatrix} \alpha_0 & \sqrt{\beta_1} & & & 0 \\ \sqrt{\beta_1} & \alpha_1 & \sqrt{\beta_2} & & \\ & \sqrt{\beta_2} & \alpha_2 & \sqrt{\beta_3} & \\ & & \ddots & \ddots & \ddots \\ 0 & & & & \end{bmatrix} \tag{4.31}$$

and its $n \times n$ leading principal minor matrix by

$$J_n = [J(\mu)]_{[1:n,1:n]}. \tag{4.32}$$

Setting $\vec{p}_n(s) = [\tilde{p}_0(s), \tilde{p}_1(s), \ldots, \tilde{p}_{n-1}(s)]^T$, equation (4.28) may be expressed in matrix form as [57]

$$J_n\vec{p}_n(\lambda) = \lambda\vec{p}_n(\lambda) - \sqrt{\beta_n}\tilde{p}_n(\lambda)\vec{e}_n, \tag{4.33}$$

where $\vec{e}_n = [0, 0, \ldots, 1]$ is the nth standard basis vector in $\mathbb{R}^n$. Therefore, the simple zeros $\{\lambda_{j,n}\}_{j=1}^n$ of $\tilde{p}_n$ are the eigenvalues of the Jacobi matrix J_n, and the $\{\vec{p}_n(\lambda_{i,n})\}_{i=1}^n$, are the corresponding eigenvectors:

$$J_n\vec{v}_i = \lambda_{i,n}\vec{v}_i, \quad \vec{v}_i^{\,T}\vec{v}_i = 1, \quad \vec{v}_i = \frac{\vec{p}_n(\lambda_{i,n})}{\|\vec{p}_n(\lambda_{i,n})\|}, \quad \|\vec{p}_n(\lambda_{i,n})\|^2 = \sum_{j=0}^{n-1}[\tilde{p}_j(\lambda_{i,n})]^2. \tag{4.34}$$

As $\Sigma_\mu \subset [0, 1]$, the coefficients of the Jacobi matrix satisfy the following bounds [57]

$$0 < \alpha_j < 1, \quad 0 < \beta_j \leq 1. \tag{4.35}$$

Therefore, the matrix $J(\mu)$ is a *bounded* Jacobi matrix, which in turn implies the measure of orthogonality μ uniquely defines $J(\mu)$ [73].

We now construct the Jacobi matrix $J_n(\mu)$ for finite lattices, where the spectral measure μ of the matrix M_n is given by equations (4.19)–(4.21). In general, given a real symmetric matrix A, there is an orthogonal similarity transformation $Q^T AQ = T$, where the orthogonal matrix Q and the tridiagonal symmetric matrix T, having nonnegative off diagonal elements, are uniquely determined by A and the first column of Q [96]. Theorem 7 explicitly determines Q and implicitly determines J_n. Although, the proof thereof defines a numerical algorithm for determining J_n. Parts of this proof may be found in separate discussions given in [57]. We provide a complete proof here.

Theorem 7 *Let* $\mu(d\lambda) = \sum_{j=1}^n m_j\delta_{\lambda_j}(d\lambda)$ *be the spectral measure of* M_n *given in* (4.21), *and define the diagonal matrix* $\Lambda_n = diag[\lambda_j]_{j=1}^n$ *and the vectors* $\vec{m} = [m_1, m_2, \ldots, m_n]^T$ *and*

$\sqrt{\vec{m}} = [\sqrt{m_1}, \sqrt{m_2}, \ldots, \sqrt{m_n}]^T$. Furthermore, let $J_n(\mu)$ be the Jacobi matrix of order n defined by equations (4.31)–(4.34) with eigenvalues $\{\lambda_{j,n}\}_{j=1}^n$ and eigenvectors $\{\vec{p}_n(\lambda_{j,n})\}_{j=1}^n$, where $\vec{p}_n(s) = [\tilde{p}_0(s), \tilde{p}_1(s), \ldots, \tilde{p}_{n-1}(s)]^T$ and $\{\tilde{p}_j\}_{j=0}^{n-1}$ are the polynomials which are orthonormal in $L^2(\mu)$. Denote by $\vec{P} = [\vec{p}_n(\lambda_{1,n}), \vec{p}_n(\lambda_{2,n}), \ldots, \vec{p}_n(\lambda_{n,n})]$ the matrix of orthogonal eigenvectors with norms $\xi_i = \|\vec{p}_n(\lambda_{i,n})\|$, and $V_n = [\vec{v}_1, \vec{v}_2, \ldots, \vec{v}_n]$ the corresponding matrix of orthonormal eigenvectors $\vec{v}_i = \vec{p}_n(\lambda_{i,n})/\|\vec{p}_n(\lambda_{i,n})\|$. Then there exists a unique orthogonal matrix Q_n of order n and a unique tridiagonal symmetric matrix T_n, with nonnegative off diagonal elements, given by

$$\begin{bmatrix} 1 & \vec{0}^T \\ \vec{0} & Q_n^T \end{bmatrix} \begin{bmatrix} 1 & \sqrt{\vec{m}}^T \\ \sqrt{\vec{m}} & \Lambda_n \end{bmatrix} \begin{bmatrix} 1 & \vec{0}^T \\ \vec{0} & Q_n \end{bmatrix} = \begin{bmatrix} 1 & \sqrt{\vec{m}}^T Q_n \\ Q_n^T \sqrt{\vec{m}} & Q_n^T \Lambda_n Q_n \end{bmatrix} = \begin{bmatrix} 1 & \sqrt{\beta_0}\vec{e}_1^T \\ \sqrt{\beta_0}\vec{e}_1 & T_n \end{bmatrix}, \tag{4.36}$$

where $\vec{e}_1$ and $\vec{0}$ are the 1st standard basis and null vectors of $\mathbb{R}^n$, respectively, and the constant $\beta_0 = \mu_0$, the mass of μ, depends on n through this measure. Furthermore, $Q_n \equiv V_n^T$, $T_n \equiv J_n(\mu)$, and $m_i = \xi_i^{-2}$.

Proof: Define a real symmetric matrix A of order $n+1$ by

$$A = \begin{bmatrix} 1 & \sqrt{\vec{m}}^T \\ \sqrt{\vec{m}} & \Lambda_n \end{bmatrix}. \tag{4.37}$$

As mentioned above, there exists [96] a similarity transformation $Q^T A Q = T$, where the orthogonal matrix Q and the tridiagonal symmetric matrix T, having nonnegative off diagonal elements, are uniquely determined by A and the first column of Q. Set the first column of Q to $\vec{e}_1$:

$$Q = \begin{bmatrix} 1 & \vec{0}^T \\ \vec{0} & Q_n \end{bmatrix}. \tag{4.38}$$

Therefore, the orthogonal matrix Q_n of order n and the tridiagonal matrix T of order $n+1$ are unique [96].

By construction, as T is tridiagonal,

$$T = Q^T A Q = \begin{bmatrix} 1 & \sqrt{\vec{m}}^T Q_n \\ Q_n^T \sqrt{\vec{m}} & Q_n^T \Lambda_n Q_n \end{bmatrix} = \begin{bmatrix} 1 & c\vec{e}_1^T \\ c\vec{e}_1 & T_n \end{bmatrix}, \tag{4.39}$$

where T_n is a unique tridiagonal symmetric matrix with nonnegative off diagonal elements and $c \geq 0$ is a constant. Therefore $T_n = Q_n^T \Lambda_n Q_n$ and Q_n is a unique orthogonal matrix. In the discussion directly below equation (4.22) we already established that

$\{\lambda_{j,n}\} \equiv \{\lambda_j\}$. Therefore by definition, $J_n(\mu) = V_n^T \Lambda_n V_n$, whereby ordering the simple eigenvalues, $\lambda_{1,n} < \lambda_{2,n} < \cdots < \lambda_{n,n}$, V_n is a unique orthogonal matrix and $J_n(\mu)$ is a tridiagonal symmetric matrix with nonnegative off diagonal elements. Therefore we have that $Q_n T_n Q_n^T = V_n J_n(\mu) V_n^T$ where Q_n, T_n, and V_n are unique, which implies that $V_n \equiv Q_n^T$ and $T_n \equiv J_n(\mu)$.

We now show that $c = \sqrt{\beta_0}$, i.e. $V_n\sqrt{\vec{m}} = \sqrt{\beta_0}\,\vec{e}_1$. Recalling that the mass μ_0 of the measure μ is $\beta_0 = p_1$ and from (4.28) we have $\tilde{p}_0 = \beta_0^{-1/2}$, by orthonormality we have $\beta_0^{1/2}\delta_{0,l} = \langle 1, \tilde{p}_l\rangle_\mu = \sum_{j=1}^n m_j \tilde{p}_l(\lambda_{j,n})$, $l = 0, 1, \ldots, n-1$, or in matrix form

$$\vec{P}\vec{m} = \beta_0^{1/2}\vec{e}_1, \tag{4.40}$$

with $\vec{P}$ and $\vec{m}$ defined in the statement of the theorem. Therefore if we define the diagonal matrix $\Xi = \mathrm{diag}[\xi_1, \xi_2, \ldots, \xi_n]$, with $\xi_i = \|\vec{p}_n(\lambda_{i,n})\|$, and $\vec{1} = [1, 1, \ldots, 1]^T \in \mathbb{R}^n$, then multiplying (4.40) from the left by $\vec{P}^T$ we obtain

$$\Xi^2\vec{m} = \beta_0^{1/2}\vec{P}^T\vec{e}_1 = \beta_0^{1/2}\beta_0^{-1/2}\vec{1} = \vec{1} \tag{4.41}$$

From (4.41) we recover the Christoffel numbers m_j of M_n in (4.30) from the norms of the eigenvectors of J_n: $\vec{m} = \Xi^{-2}\vec{1}$, or in coordinate form

$$m_i = \xi_i^{-2}, \qquad \xi_i = \|\vec{p}_n(\lambda_{i,n})\|, \quad i = 1, 2, \ldots, n. \tag{4.42}$$

Equivalent to (4.42) is $\sqrt{m_i} = \|\vec{p}_n(\lambda_{i,n})\|^{-1}$. Therefore, recalling that the first component of the vectors $v_{i,1}$ are given by $v_{i,1} = \beta_0^{-1/2}\|\vec{p}_n(\lambda_{i,n})\|^{-1}$, we have $\sqrt{m_i} = \beta_0^{1/2}v_{i,1}$. Or in matrix form $\sqrt{\vec{m}}^{\,T} = \sqrt{\beta_0}\vec{e}_1^{\,T}V$, where $\sqrt{\vec{m}}^{\,T}$ is defined in the statement of the theorem. Therefore, by the orthogonality of the matrix V, this expression yields

$$V\sqrt{\vec{m}} = \sqrt{\beta_0}\vec{e}_1, \tag{4.43}$$

which concludes the proof of Theorem 7 □.

The procedure leading to equation (4.36) defines a *Lanczos*-type algorithm. Given the spectral parameters $\{m_j, \lambda_j\}$ of M_n, which define the matrix A, a stable variant of this algorithm [57] accurately produces Q and T. The matrix T is the "extended" Jacobi matrix on the far right of equation (4.36). The nonzero elements of the submatrix J_n are the recursion coefficients, $\{\alpha_{j,n}\}_{j=0}^n$ and $\{\sqrt{\beta_{j,n}}\}_{j=1}^n$, with $\beta_0 = \sum_{j=1}^n m_j = p_1$ [57].

In Figure 4.1, we give a graphical representation of the eigenvalue densities $\rho(\lambda)$ for the matrix $\chi_2\Gamma\chi_2$ associated with 2-d and 3-d RRN with i.i.d. bonds. These eigenvalue

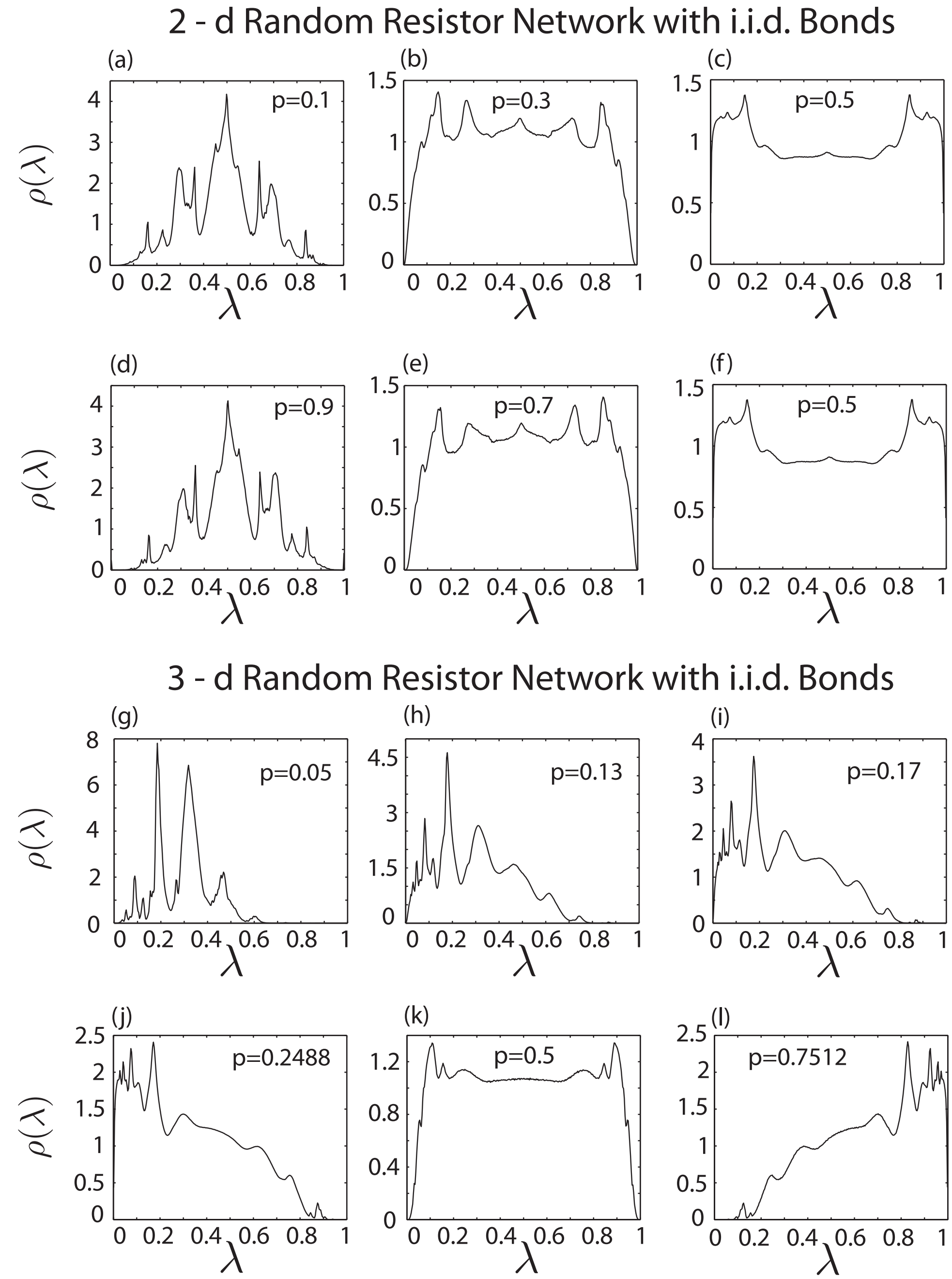

Figure 4.1. In the 2-d RRN (a)–(c), as the volume fraction p increases from left to right, the width of the gaps in the spectrum near $\lambda = 0, 1$ shrink to 0, *symmetrically* with increasing connectedness as $p \longrightarrow p_c = 1 - p_c = 0.5$. Displayed directly below each of the panels (a)–(c) for volume fraction p, are the corresponding eigenvalue densities (d)–(f) for volume fraction $1-p$, illustrating the symmetry $\rho(p,\lambda) = \rho(1-p, 1-\lambda)$. In the 3-d RRN (g)–(l), as $p \longrightarrow p_c \approx 0.2488$ the width of the gap near $\lambda = 0$ shrinks to 0, and as $p \longrightarrow 1 - p_c \approx 0.7512$ the width of the gap near $\lambda = 1$ shrinks to 0. These eigenvalue densities also have the symmetry $\rho(p,\lambda) = \rho(1-p, 1-\lambda)$.

densities are precisely that of the Jacobi matrix associated with $\chi_2\Gamma\chi_2$, as they have the same eigenvalues, which we have numerically verified using the above mentioned Lanczos algorithm [57]. These figures display linearly connected peaks of histograms with bin sizes on the order of 10^{-2}. The apparent smoothness of these graphs in is due to the large number ($\sim 10^6$) of eigenvalues calculated, and ensemble averaged. These figures illustrate the symmetry $\rho(p,\lambda)=\rho(1-p,1-\lambda)$ in the *bulk* of the spectrum, where $p=p_2$. This follows from the invariance of the binary model, under the simultaneous interchange $p\leftrightarrow 1-p$, $\sigma_1\leftrightarrow\sigma_2$ [39, 65]. As p surpasses the percolation threshold p_c, the gaps in the spectrum near $\lambda=0,1$ collapse and the spectra subsequently pile up into δ function-like singularities at $\lambda=0,1$.

Let us review what we have established so far. Given an infinite stationary two-phase random medium, the spectral theorem [101] applied to the associated bounded self-adjoint operator $M=\chi_1\Gamma\chi_1$ provides us with a positive measure μ with compact support $\Sigma_\mu\subseteq[0,1]$. This measure defines an infinite sequence of orthogonal polynomials which, in turn defines a bounded essentially self adjoint Jacobi matrix J of infinite order. There is, in fact, a one-to-one mapping between bounded Jacobi matrices of infinite order and positive measures with compact support. In the matrix case this map becomes a bijection and the spectral measure for J_n is parameterized by $2n$ real numbers (see Theorem 7):

$$\left\{(\lambda_1<\lambda_2<\cdots<\lambda_n,\xi_1^{-1},\xi_2^{-1},\dots,\xi_n^{-1}):\ \xi_i>0,\ \sum_{j=1}^{n}\left(\xi_j^{-1}\right)^2=p_1\right\}. \tag{4.44}$$

These results are summarized in Theorem 8 below [41]. For simplicity, in this theorem we consider measures with unit mass, e.g. $\mu\mapsto\mu/p_1$. To characterize the mapping between positive probability measures and Jacobi operators for the finite matrix case, we define

$$\mathscr{A}_1=\left\{(\gamma_1<\gamma_2<\cdots<\gamma_n,\theta_1,\theta_2,\dots,\theta_n)|\ \theta_i>0,\ \sum_{j=1}^{n}\theta_j^2=1\right\}. \tag{4.45}$$

Theorem 8 (Spectral Theorem for Jacobi Matrices) *Denote by $\mathcal{B}_J$ and $\mathcal{B}_{J_n}$ the set of bounded Jacobi matrices of infinite order and order n, respectively. Moreover, denote by $\mathscr{B}$ the set of positive Borel measures on $\mathbb{R}$ with compact support and denote $\mathcal{M}_1=\{\tau\in\mathscr{B}|\int d\tau=1\}$. There exists a one-to-one map $\psi=\varphi^{-1}$, $\psi:\mathcal{M}_1\mapsto\mathcal{B}_J$, such that for all $\tau\in\mathcal{M}_1$ there exists $J\in\mathcal{B}_J$ such that $\tau=\varphi(J)=\varphi\circ\psi(\tau)$. Furthermore, for all $J\in\mathcal{B}_J$ there exists a unique measure $\tau\in\mathcal{M}_1$ such that*

$$\langle (s-J)^{-1}\vec{e_0}, \vec{e_0}\rangle = \int \frac{d\tau(\lambda)}{s-\lambda} \tag{4.46}$$

Moreover, $J \overset{\varphi}{\mapsto} d\tau$ is the spectral map in the sense that the map U defined by

$$l_2 \ni \pi(J)\vec{e_0} \overset{U}{\mapsto} \pi(\lambda) \in L^2(\tau), \quad \pi \text{ a polynomial}, \tag{4.47}$$

extends to a unitary map and

$$(UJU^{-1}f)(\lambda) = \lambda f(\lambda), \quad \forall f \in L^2(\tau). \tag{4.48}$$

In the matrix case, the spectral map

$$\mathcal{B}_{J_n} \overset{\varphi}{\mapsto} \tau = \sum_{j=1}^{n} \xi_j^{-2}\delta_{\lambda_j} \mapsto (\lambda_1, \lambda_2, \cdots, \lambda_n, \xi_1^{-1}, \xi_2^{-1}, \dots, \xi_n^{-1}) \in \mathscr{A}_1$$

is a bijection, where the weights $\xi_i = \|\vec{p}_n(\lambda_i)\|$ are defined in Theorem 4.36 and $\mathscr{A}_1$ is defined in (4.45). In particular, for fixed $\Lambda_0 = diag(\lambda_{10}, \lambda_{20}, \dots, \lambda_{n0})$, the isospectral set $\mathscr{M}_{\Lambda_0}$, where

$$\mathscr{M}_{\Lambda_0} = \{J \in \mathcal{B}_J : spec\ J = (\lambda_{10}, \lambda_{20}, \dots, \lambda_{n0})\},$$

is the positive n-tant of the unit sphere S_1^{n-1} in $\mathbb{R}^n$.

For a proof of Theorem 8 see [41]. Geometrically speaking, in the infinite order case, the spectral map $J \overset{\varphi}{\mapsto} \tau$ takes $\mathcal{B}_J$ into the hyperplane $\mathcal{M}_1 \subset \mathscr{B}$ [41]. More specifically, given $J \in \mathcal{B}_J$, one considers the isospectral set $\mathscr{M}_0 = \{J_0 \in \mathcal{B}_J : \text{spec } J_0 = \text{spec } J\}$. Under φ, the set $\mathscr{M}_0$ is injected into the hyperplane $\mathcal{M}_1$ [41]. The geometry of this injection, i.e., how $\varphi(\mathscr{M}_0)$ lies inside $\mathcal{M}_1$, can be conveniently described in terms of basic notations of real analysis [41, 44].

We conclude this section by summarize the mapping structure associated with two-phase stationary random media. Such a medium is determined by a probability space (Ω, P), introduced in Section 2.1. Denote by $\mathcal{B}_\times$ the class of operators with domain $\mathscr{H}_\times \subset L^2(\Omega, P)$ that are: bounded, linear, self-adjoint, and have simple spectrum, where $\mathscr{H}_\times$ is defined in equation (2.1). For each $\omega \in \Omega$, the resolvent representation of the electric field in (2.10) defines a map $\psi_\omega : \mathscr{H}_\times \mapsto \mathcal{B}_\times \ni M = \chi_1\Gamma\chi_1$. The spectral theorem [101] then provides a map $\varphi_\mu : \mathcal{B}_\times \mapsto \mathcal{M}_{p_1} \ni \mu = \langle E_\lambda \vec{e}_0, \vec{e}_0\rangle$, where $\langle\cdot,\cdot\rangle$ is the $L^2(\Omega, P)$ inner product weighted by χ_1, E_λ is the resolution of the identity associated with the operator M, and $\vec{e}_0$ is a "cyclic vector" [101]. The one-to-one map $\psi : \mathcal{M}_{p_1} \mapsto \mathcal{B}_J$ defines a bounded Jacobi matrix $J(\mu)$ of infinite order, with orthogonality measure μ, which acts on l_2. The map $\varphi = \psi^{-1}$, extends to a unitary map $U : l_2 \mapsto \mathbb{P} \subset L^2(\mu)$, where $\mathbb{P}$ denotes the set of real polynomials on $[0,1]$. Pictorially, we have

$$(\mathscr{H}_\times \subset L^2(\Omega,P)) \xrightarrow{\psi_\omega} (M \in \mathcal{B}_\times) \xrightarrow{\varphi_\mu} (\mu \in \mathcal{M}_{p_1}) \underset{\varphi}{\overset{\psi}{\longleftrightarrow}} (J(\mu) \in \mathcal{B}_J) \underset{U^{-1}}{\overset{U}{\longleftrightarrow}} (\pi(s) \in L^2(\mu)).$$

We note that every two-phase stationary random medium (Ω, P) defines a (bounded) random operator $M \in \mathcal{B}_\times$, and there is a one-to-one correspondence [101] between M and the projection valued measures E_λ. Moreover, each "cyclic vector" [101] $\vec{e}_0 \in \mathscr{H}_\times$ gives rise to a positive spectral measure $\mu = \langle E_\lambda \vec{e}_0, \vec{e}_0 \rangle$ such that $\mu \in \mathcal{M}_{p_1}$, hence a bounded Jacobi matrix. However, the measure μ is not uniquely determined by E_λ and $\vec{e}_0$ [101] and it is known [65] that not every measure $\mu \in \mathscr{B}(\mu_1, \mu_2, \ldots, \mu_n) \subset \mathcal{M}_{p_1}$ gives rise to a function $m(h) = 1 - F(s;\mu)$ that is the effective (relative) dielectric constant of a random medium for $d > 2$ [87], where $\mathscr{B}(\mu_1, \mu_2, \ldots, \mu_n)$ is defined in equation (4.18).

4.3 Generalized Numerator Polynomials

In this section we introduce generalized numerator polynomials $P^{[i]}$, where $i = 0, 1, \ldots$. We also derive a novel, closed form solution for the moments $\mu_j^{[i]}$ of the measures $\mu^{[i]}$ in terms of the moments μ_j of μ, for all $i \in \mathbb{N}$, $j = 0, 1, \ldots$. Finally, we make connections from the Stieltjes transforms $F^{[i]}(s; \mu^{[i]})$ of the orthogonality measures $\mu^{[i]}$ underlying these families of polynomials, to the energy components of Theorem 4.

The generalized monic numerator polynomials of order $i = 0, 1, 2, \ldots$ are defined to be the solution of the three-term recursion relation [57, 124]

$$\begin{aligned} P_{n+1}^{[i]}(s) &= (s - \alpha_{n+i}) P_n^{[i]}(s) - \beta_{n+i} P_{n-1}^{[i]}(s), \quad n = 0, 1, 2, \ldots \\ P_{-1}^{[i]}(s) &= 0, \quad P_0^{[i]}(s) = 1, \end{aligned} \tag{4.49}$$

where the recursion coefficients $\{\alpha_j, \beta_j\}$ are defined in equation (4.29) and, $P_n = P_n^{[0]}$ and $Q_{n-1} = P_{n-1}^{[1]}$ are the monic denominator and numerator polynomials arising in the Padé approximants of $F(s) = F(s;\mu)$ in (4.5), respectively [57, 124]. It is an elementary exercise to show that a sequence of orthogonal polynomials satisfies a three-term recursion relation [41, 57, 73]. The converse is known as Favard's Theorem [57], which states: if an infinite sequence of polynomials satisfy a three-term recurrence relation, like that in (4.49) with $\beta_k > 0$ and $\alpha_k \in \mathbb{R}$, then there exists an orthogonality measure for this sequence.

Even though the orthogonality measures $\mu^{[i]}$ associated with the $P_n^{[i]}$ in (4.49) are not known in general, their Stieltjes transforms $F^{[i]}(s) = F^{[i]}(s;\mu^{[i]})$ can be expressed in terms of Cauchy integrals of μ (see equation 3.7 in [124]):

$$F^{[i]}(s) = \int_{\mathbb{R}} \frac{d\mu^{[i]}(\lambda)}{s-\lambda} = \frac{1}{\beta_i}\frac{\rho_i(s)}{\rho_{i-1}(s)}, \quad \rho_i(s) = \int_{\mathbb{R}} \frac{P_i(\lambda)d\mu(\lambda)}{s-\lambda}, \quad \rho_{-1}(s) = 1, \tag{4.50}$$

for $s \in \mathbb{C}\backslash\text{co}(\text{supp}(\mu))$, where $\text{co}(\text{supp}(\mu))$ is a compact interval defined in [35]. Define the functions $F_j(s)$, $j = 0, 1, \ldots$, as in equation (4.51) below. If we define the polynomials $P_i(s) = \sum_{j=0}^{i} a_j s^j$ and $P_{i-1}(s) = \sum_{j=0}^{i-1} b_j s^j$, then we may write $\rho_i(s) = \sum_{j=0}^{i} a_j F_j(s)$ and $\rho_{i-1}(s) = \sum_{j=0}^{i-1} b_j F_j(s)$. Therefore equation (4.50) may be written as

$$\beta_i \left[\sum_{j=0}^{i-1} b_j F_j(s)\right] F^{[i]}(s) = \sum_{j=0}^{i} a_j F_j(s), \quad F_j(s) = \int_0^1 \frac{\lambda^j d\mu(\lambda)}{s-\lambda}. \tag{4.51}$$

We prove in Theorem 9 below that equation (4.51) leads to a closed form solution for the moments $\mu_j^{[i]} = \int_0^1 \lambda^j d\mu^{[i]}(\lambda)$ of $\mu^{[i]}$, in terms of the moments μ_j of μ. This solution uniquely determines the measures $\mu^{[i]}$. More specifically, as μ is compactly supported, $\Sigma_\mu \subset [0,1]$, the supports of the measures $\mu^{[i]}$ are compact intervals, for all $i \geq 0$ [35]. Therefore the moment problem for the measures $\mu^{[i]}$ is determined, i.e., the $\mu^{[i]}$ are uniquely determined by their moments $\{\mu_j^{[i]}\}_{j=0}^{\infty}$ [114]. It is worth noting that the orthogonality measures $\mu^{[i]}$ may be found using the spectral resolution of the corresponding essentially self-adjoint Jacobi operator defined by (4.49) (see Theorem 10.23 page 531 in [118]). Furthermore, by equation (4.51), the measures $\mu^{[i]}$ may also be found in terms of the measures $\lambda^j \mu(d\lambda)$, for $j = 0, 1, 2, \ldots, i$, via the Stieltjes–Perron Inversion Theorem in (2.34) (see the formula after equation 3.7 in [124]).

Theorem 9 *Let $\mu_j^{[i]}$ and μ_j denote the moments of the measures $\mu^{[i]}$ and μ, respectively, and $P_n = P_n^{[0]}$ be the polynomials in (4.49). Then the $\mu_j^{[i]}$ are defined recursively by*

$$\mu_j^{[i]} = \frac{1}{\langle \lambda^{i-1}, P_{i-1}\rangle_\mu} \left[\frac{\langle \lambda^{i+j}, P_i\rangle_\mu}{\beta_i} - \sum_{l=0}^{j-1} \langle \lambda^{(i+j-1)-l}, P_{i-1}\rangle_\mu \, \mu_l^{[i]}\right] \tag{4.52}$$

for all $i = 1, 2, \ldots$ and $j = 0, 1, \ldots$, where $\langle \cdot, \cdot \rangle_\mu$ is the $L^2(\mu)$ inner-product and empty sums are understood to be zero.

Proof: By applying the expansion (4.1) to the $F_j(s)$ in equation (4.51) and $F^{[i]}(s)$ in (4.50), and switching the order of the infinite and finite sums, we have

$$\beta_i \left[\sum_{l=i-1}^{\infty} \frac{\langle \lambda^l, P_{i-1} \rangle_\mu}{s^{l+1}} \right] \left[\sum_{l=0}^{\infty} \frac{\mu_l^{[i]}}{s^{l+1}} \right] = \sum_{l=i}^{\infty} \frac{\langle \lambda^l, P_i \rangle_\mu}{s^{l+1}}, \tag{4.53}$$

where $\langle \cdot, \cdot \rangle_\mu$ denotes the $L^2(\mu)$ inner product. The change in the lower sum indices in equation (4.53) follows from the orthogonality of the P_j in $L^2(\mu)$. The moments $\mu_j^{[i]}$ of the measures $\mu^{[i]}$ may now be found by equating the coefficients of powers of s^{-1} in equation (4.53). Doing so defines an infinite (linear) system equations, $\mathbf{L}_i \vec{x} = \vec{b}$, where $x_j = \mu_j^{[i]}$ and $b_j = \langle \lambda^{i+j}, P_i \rangle_\mu / \beta_i$, for $j = 0, 1, 2, \ldots$, and $\mathbf{L}_i$ is a lower triangular Toeplitz convolution matrix:

$$\begin{bmatrix} \langle \lambda^{i-1}, P_{i-1} \rangle_\mu & 0 & 0 & 0 & \cdots \\ \langle \lambda^{i}, P_{i-1} \rangle_\mu & \langle \lambda^{i-1}, P_{i-1} \rangle_\mu & 0 & 0 & \cdots \\ \langle \lambda^{i+1}, P_{i-1} \rangle_\mu & \langle \lambda^{i}, P_{i-1} \rangle_\mu & \langle \lambda^{i-1}, P_{i-1} \rangle_\mu & 0 & \cdots \\ \vdots & \vdots & \vdots & \ddots & \cdots \end{bmatrix} \begin{bmatrix} \mu_0^{[i]} \\ \mu_1^{[i]} \\ \mu_2^{[i]} \\ \vdots \end{bmatrix} = \begin{bmatrix} \langle \lambda^{i}, P_i \rangle_\mu / \beta_i \\ \langle \lambda^{i+1}, P_i \rangle_\mu / \beta_i \\ \langle \lambda^{i+2}, P_i \rangle_\mu / \beta_i \\ \vdots \end{bmatrix}. \tag{4.54}$$

Therefore the moments of the measure $\mu^{[i]}$ may be found by back substitution, and are given recursively by equation (4.52). This concludes the proof of Theorem 9 □.

We now show that the $F^{[i]}(s)$, for $i = 0, 1, 2$, are completely determined by energy components of the decomposition given in Theorem 4. From equation (4.51) we see that $F^{[0]}(s) = F(s)/p_1$, and $F^{[1]}(s)$ and $F^{[2]}(s)$ are given in terms of the functions $F_1(s)$ and $F_2(s)$ defined in equation (4.51). Theorem 4 shows that

$$sF(s) = \langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle / E_0^2, \quad F_1(s) = \langle \chi_1 \vec{E}_f \cdot \vec{E}_0 \rangle / E_0^2, \quad F_2(s) = -\int_\infty^s \langle \chi_1 E_f^2 \rangle / E_0 \, ds, \tag{4.55}$$

where $\lim_{|s| \longrightarrow \infty} F_2(s) = 0$ and $\langle \cdot \rangle$ denotes the averaging defined in Section 2.1. Equation (4.49) with $i = 0$ implies that $P_0(\lambda) = 1$, $P_1(\lambda) = \lambda - \alpha_0$, and $P_2(\lambda) = \lambda^2 - (\alpha_0 + \alpha_1)\lambda + \alpha_0\alpha_1 - \beta_1$. Therefore equations (4.51) and (4.55) yield [35]

$$\begin{aligned} F^{[1]}(s) &= \frac{\langle \chi_1 \vec{E}_f \cdot \vec{E}_0 \rangle - (\alpha_0/s) \langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle}{(\beta_1/s) \langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle}, \\ F^{[2]}(s) &= \frac{-\int_\infty^s ds \langle \chi_1 E_f^2 \rangle - (\alpha_0 + \alpha_1) \langle \chi_1 \vec{E}_f \cdot \vec{E}_0 \rangle + [(\alpha_0\alpha_1 - \beta_1)/s] \langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle}{\beta_2 (\langle \chi_1 \vec{E}_f \cdot \vec{E}_0 \rangle - (\alpha_0/s) \langle \chi_1 \vec{E} \cdot \vec{E}_0 \rangle)}. \end{aligned} \tag{4.56}$$

Clearly, for all $i \geq 0$, the $F^{[i]}(s)$ may be expressed in terms of the $F_j(s)$, for $j = 0, 1, \ldots, i$, and therefore may be expressed in terms of the energy components in (4.55). However, for $i = 0, 1, 2$ the $F^{[i]}(s)$ are given *completely* in terms of these energy components.

Equations (4.50), (4.51), (4.55), and (4.56) are general formulas that hold for two-component stationary random media in the lattice and continuum settings [60], and hold for all $s \in \mathbb{C}\backslash \mathrm{co}(\mathrm{supp}(\mu))$ [35]. It is interesting to note that, as the roots of $P_n^{[1]}$ interlace those of P_n and the roots of $P_n^{[2]}$ interlace those of $P_n^{[1]}$ etc. [118], the root densities $\rho^{[i]}(\lambda)$ of these polynomials are very similar to that shown in Figure 4.1. We have calculated histogram representations of $\rho^{[i]}(\lambda)$ for several values of i using the above mentioned Lanczos algorithm [57] and, even though the roots of these polynomials are different, the graphs of these histograms look identical.

CHAPTER 5

RANDOM MATRIX THEORY FOR COMPOSITES

In Section 2.1 we introduced four self-adjoint random operators $M_j = \chi_j \Gamma \chi_j$ and $K_j = \chi_j \Upsilon \chi_j$, $j = 1, 2$, which are at the heart of Stieltjes integral representations [11, 65, 86] of the effective conductivity and resistivity tensors $\boldsymbol{\sigma}^*$ and $\boldsymbol{\rho}^*$, respectively. In the lattice setting of a *finite* RRN, these random operators are real-symmetric random matrices [58, 64, 89]. In this chapter, we demonstrate that, for percolation models of RRN [60, 89, 116, 122], the eigenvalue statistics of the random matrices $M_j = \chi_j \Gamma \chi_j$, $j = 1, 2$, exhibit a transitional behavior like that of a universality class of q-deformed random matrix ensembles (RME) [93, 94]. We will focus on the operator M_2 as the results associated with the operator M_1 follow by the symmetry $\chi_1 = 1 - \chi_2$.

5.1 q-Random Matrix Ensembles

Random matrix theory (RMT) was introduced by Wigner [129] and Dyson [47–49] to provide a statistical description of the quantized energy levels of heavy nuclei. Since then, it has been applied in studies of quantum chaos [1, 22, 67], biological networks [82, 99], random graphs [7, 77], the Anderson transition of quantum/mesoscopic conductors [28, 29, 76, 93, 94, 117], log-gases [56], and even the Riemann hypothesis of analytic number theory [15, 84, 85]. Poisson statistics of a random matrix are characterized by the *absence* of eigenvalue correlations [67]. Wigner–Dyson (WD) statistics of a random matrix are characterized by *strong* eigenvalue correlations, giving rise to the phenomenon of eigenvalue repulsion [26, 67, 117]. WD eigenvalue correlations typically exist among the eigenvalues of the Gaussian ensemble (GE) of a *real-symmetric* random matrix H, that is, an ensemble of matrices randomly distributed with probability measure [6, 41, 42, 84, 97]

$$P_n[H]d[H] = \tilde{Z}_n^{-1} \exp[-\beta \, \mathrm{Tr} H^2] d[H], \quad d[H] = \prod_{k \geq j} dH_{kj} \tag{5.1}$$

defined on the space of $n \times n$ real-symmetric matrices, where $\beta = 1$, $\tilde{Z}_n$ is the normalization factor, $d[H]$ is the "Lebesgue" measure for real-symmetric matrices [6], and Tr denotes matrix trace. The GE of a *real-symmetric* random matrix is called the Gaussian orthogonal ensemble (GOE). WD eigenvalue correlations also arise in GEs of complex Hermitian and quaternion self-dual random matrices [84], and they are called the Gaussian unitary ensemble (GUE) and the Gaussian symplectic ensemble (GSE), respectively. In equation (5.1), $\beta = 2$ for the GUE and $\beta = 4$ for the GSE. The random matrices that we consider here are real-symmetric, and we will henceforth focus on the GOE and its variants.

The GEs, and the associated WD statistics, do not bear any hint of the spatial dimensionality d of a physical system, and are parameter independent [26]. Furthermore, by definition, they are invariant under similarity transformations [42] and thus have no basis preference in them [26]. This means [26] that they describe systems where (1) all the normalized linear combinations of the eigenstates have similar properties and (2) the dimensionality, in some sense, is irrelevant.

Early on, numerical simulations demonstrated that GEs with a modified probability density also exhibit WD statistics: $P_n(H) \propto \exp[-\mathrm{Tr}V(H)]$, with V a real valued bounded below function satisfying $V(\lambda) \sim |\lambda|^a$ for $a \geq 1$, as $|\lambda| \longrightarrow \infty$ [26, 84, 97]. More specifically, the mean density of eigenvalues ρ is highly sensitive to the form of the function V, although the fluctuations about ρ are *universal*. Recently, universality in the bulk, and at the edge of the spectrum has been established for V in the class of even ordered polynomials [41–43].

Remarkably, when one changes variables from matrix elements H_{jk} to eigenvalues $\{\lambda_l\}$ and eigenvectors $\{\vec{q}_l\}$, with $H = \sum_{l=1}^{n} \lambda_l \vec{q}_l \vec{q}_l^{\,T}$, the probability density in (5.1) becomes exactly that [41, 43, 97] of the canonical ensemble of n unit charges on a line at temperature $\beta^{-1} = 1$, confined by a potential V, and repelling one another logarithmically:

$$P_n[H]d[H] = Z_n^{-1} \exp(-\beta \mathcal{H}_n) d\lambda_1 \cdots d\lambda_n dQ, \quad \mathcal{H}_n = \beta^{-1} \sum_{i=1}^{n} V(\lambda_i) - \sum_{i>j=1}^{n} \ln |\lambda_i - \lambda_j|, \tag{5.2}$$

where $\mathcal{H}_n$ is the energy Hamiltonian, Z_n is the normalization factor, and dQ is the Haar measure on $Q(n)$, the group of orthogonal transformations on $\mathbb{R}^{n\times n}$ [42]. The logarithmic term in (5.2) comes from the Jacobian of the variable change, and is *independent* of the choice of the function V [6, 26, 42]. Equation (5.2) demonstrates that the eigenvalues and eigenvectors of these RMEs are statistically independent, and that the eigenvectors are always distributed "uniformly" over the part of the orthogonal group $Q(n)$ specified by $\{\vec{q}_l =$

$(q_{1l}, \ldots, q_{nl}),\ q_{1l} \geq 0\}_{l=1}^{n}$ [97]. Integrating over eigenvectors, and absorbing the resultant constant in the normalization Z_n, yields a theory of only eigenvalues.

In total agreement with statistical mechanics [50, 97], for functions V satisfying a Lipschitz type condition, and having the asymptotic behavior $V(\lambda) \gtrsim (2+\epsilon)\ln|\lambda|$ as $|\lambda| \longrightarrow \infty$, where $\epsilon > 0$, the ground state distribution of the eigenvalues is given by the minimum "electrostatic energy" whose distribution is given by the unique equilibrium measure ρ of the associated infinite volume free energy: $f[\rho] = \inf_m f[m] = \lim_{n\longrightarrow\infty} n^{-2}\ln Z_n$. Here

$$f[m] = \int \left(V(\lambda) - \frac{\beta}{2}U(\lambda; m)\right) dm(\lambda), \quad U(\lambda; m) = \int \ln|\lambda - \lambda'| dm(\lambda'), \tag{5.3}$$

is varied over eigenvalue distributions described by the measure m [26, 97]. We stress that the corresponding variational problem determines both the (compact) support and the form of the measure ρ [97].

In [6] Balian demonstrated that this connection from RMT to statistical mechanics is even deeper, by proving that the form of the probability density in (5.2) follows from maximizing an entropy functional $S\{P[H]\} = \int d[H]P[H]\ln P[H]$. The function V acts like a generalized Lagrange multiplier, which is determined by a constraint such as $\langle \mathrm{Tr} V(H)\rangle = C$ or, for example, by requiring that the density of eigenvalues is a given function $\rho(\lambda)$, which is taken directly from the microscopic system being investigated [6, 26, 117]: $\rho(\lambda) = \langle \mathrm{Tr}[\delta(\lambda - H)]\rangle$. This yields $V(\lambda) = \int \ln|\lambda - \lambda'|\rho(\lambda')d\lambda'$ to order $n^{-1}\ln n$ [8].

Dyson formulated [46, 50] a Brownian-motion model for the evolution of an ensemble of random matrices. He introduced a fictitious "time" τ, and modeled the τ dependence of the distribution of eigenvalues $\vec{\lambda}(\tau) = (\lambda_1(\tau), \ldots, \lambda_n(\tau))$ at temperature β^{-1}, in a fictitious, viscous fluid with friction coefficient γ. The associated probability distribution $P(\vec{\lambda}, \tau)$ of the eigenvalues evolves [8, 50] according to the Fokker–Planck equation

$$\gamma \frac{\partial P}{\partial \tau} = \vec{\nabla}_\lambda \cdot (\beta P\, \vec{\nabla}_\lambda \mathcal{H}_n + \vec{\nabla}_\lambda P), \tag{5.4}$$

where $\vec{\nabla}_\lambda$ is the gradient operator with respect to the $\{\lambda_j\}$ and $\mathcal{H}_n$ is defined in equation (5.2). The limiting solution $P(\vec{\lambda}, \tau)$ of (5.4) as $\tau \longrightarrow \infty$ is the "equilibrium" distribution $P_{eq} = Z_n^{-1}\exp(-\beta\mathcal{H}_n)$ of the eigenvalues given by equation (5.2) [8, 46, 50]. In [8], Beenakker and Rejaei have shown that one can actually identify the fictitious time τ with an external perturbation parameter X, with $\tau = X^2$. The parameter X can be, for example, an electric field or magnetic field acting on the system. This allows one to obtain various eigenvalue correlation functions at different values of X, which are usually called parametric correlations [16].

There are many examples [26] of *disordered* systems that are governed by a random matrix, which exhibits WD statistics in one disorder regime, while the statistics significantly deviate away toward Poisson-like statistics, as a function of disorder. Studies of these systems have inferred that the transition from universal WD statistics of eigenvalues toward Poisson-like statistics of eigenvalues is given by a "soft" confining potential of the form $V(\lambda) = A\ln^2|\lambda|$, $0 < A < 1$ [94]. More specifically, decreasing A causes the eigenvalues of the RME in (5.2) to transition from WD-like statistics towards Poisson-like statistics [26]. It has been suggested [26] that the Poissonian behavior of such RMEs is due to the spontaneous breakdown of the $Q(n)$ symmetry at the transition from a power-law potential $V(\lambda) \sim |\lambda|^a$, with $a \geq 1$, to the logarithmic potential $V(\lambda) = A\ln^2|\lambda|$ as $a \longrightarrow 0$, yielding RMEs with multifractal eigenvectors [76]. In [26], Canali generalized Dyson's mean field theory [50] to such RMEs, which holds in the bulk of the spectrum, and accurately describes the Anderson transition of mesoscopic/quantum conductors [117].

Using a class of potentials $V(\lambda; q)$ that depends on a disorder parameter q, this transition from WD statistics towards Poisson-like statistics has been captured by a single model [93, 94]. As the value of q varies, the asymptotic behavior of $V(\lambda; q)$ transitions from strong confinement $V(\lambda; q) \gtrsim |\lambda|$ to soft confinement $V(\lambda; q) \sim \ln^2|\lambda|$. This new *one parameter* universality class of RMEs have been called q-deformed random matrix ensembles (q-RME). By a generalization of Mehta's method of orthogonal polynomials [84], this problem has been analytically solved [93, 94] for q-deformed GUEs (q-GUE). However, the associated analytical work for the q-deformed GOE (q-GOE) and q-deformed GSE (q-GSE), are currently open problems. Dyson's Brownian-motion model in (5.4) has also been generalized to q-RMEs in [16] and solved for q-GUEs. This stochastic model should be relevant for conductors at stronger disorder, and could potentially provide details regarding the scaling regime of two-phase conductive media. In Section 5.2, we numerically demonstrate that, for percolation models of finite RRN, the eigenvalue statistics associated with the random matrix $M_2 = \chi_2\Gamma\chi_2$ transition from WD-like statistics toward Poisson-like statistics as a function of the disorder parameter p, the volume fraction of σ_2 bonds.

5.2 Random Matrix Statistics

In this section, we introduce some important eigenvalue statistics of RMT, and present the key results of this chapter. These RMT statistics were originally created by Dyson and Mehta [51] to study short and long-range correlations of the quantized energy levels of heavy nuclei. In percolation models of two-phase random media, such as RRN, these statistics provide valuable insight regarding critical connectedness transitions exhibited by such media.

Consider the real-symmetric random matrix $M_2 = \chi_2 \Gamma \chi_2$, introduced in Section 3.3, underlying the effective conductance σ^* a *finite* RRN. Each geometric configuration $\omega \in \Omega$ of the RRN determines a statistical realization of the random matrix $M_2(\omega) = \chi_2(\omega)\Gamma\chi_2(\omega)$, and an ordered sequence of eigenvalues $\vec{\lambda}(\omega) = \{\lambda_1(\omega), \ldots, \lambda_n(\omega)\}$. This family of sequences defines a mean eigenvalue density ρ. In order to analyze the fluctuation properties of the eigenvalues $\vec{\lambda}(\omega)$ about ρ, the spectrum has to be unfolded [67], i.e., the system-specific mean eigenvalue density must be removed from the data $\vec{\lambda}(\omega)$, for each $\omega \in \Omega$. The variable that serves this purpose is the integrated density of states [26]

$$u(\lambda) = \int_0^\lambda \rho(\lambda')d\lambda'. \tag{5.5}$$

This maps $\lambda_j \mapsto u_j$, for each $j = 1, \ldots, n$, and in this new variable the spectra $\{u_j\}$ has uniform spacing [26, 67], which we will normalize to have *unit mean spacing*.

The nearest neighbor spacing distribution $P(s)$ is the observable most commonly used to study short-range correlations in the spectrum, where the spacing variable s is not to be confused with the conductance contrast parameter. It is equal to the probability density for two *neighboring* eigenvalues u_n and u_{n+1} having the spacing s. For the Poisson spectrum without correlations $P(s) = \exp(-s)$, and for WD spectrum with strong correlations $P(s) \approx \frac{\pi}{2} s \exp(-\pi s^2/2)$, known as the Wigner surmise [67]. The Wigner surmise approximates the actual spacing distribution quite well and illustrates the phenomenon of eigenvalue repulsion, i.e., the probability of zero spacings is zero.

The nearest neighbor spacing distribution contains information about the spectrum which involves short scales (a few mean spacings). Long-range correlations are measured by quantities such as the eigenvalue number variance $\Sigma^2(L)$ and the spectral rigidity $\Delta_3(L)$. Let $\eta(L, u_s)$ denote the number of eigenvalues in the interval $[u_s, u_s + L]$, on the unfolded scale. The number variance is given by

$$\Sigma^2(L) = \langle \eta^2(L, u_s) \rangle - \langle \eta(L, u_s) \rangle^2, \tag{5.6}$$

where $\langle \cdot \rangle$ denotes averaging over starting points u_s. By construction, on the unfolded scale one has $\langle \eta(L, u_s) \rangle = L$. Therefore, in an interval of length L one expects, on average, $L \pm \sqrt{\Sigma^2(L)}$ eigenvalues. For the Poisson spectrum without correlations $\Sigma^2(L) = L$, and for WD spectrum $\Sigma^2(L) = (2/\pi^2)(\ln(2\pi L)+\gamma+1-\pi^2/8)$, to order $1/L$, where $\gamma = 0.5772\ldots$ is Euler's constant [67].

The spectral rigidity $\Delta_3(L)$ is closely related to $\Sigma^2(L)$. Denote the counting measure ν of a sequence of calculated eigenvalues by $\nu(u) = \sum_j \delta(u - u_j)$. The cumulative spectral function $\eta(u) = \int_{-\infty}^{u} \nu(u')du'$ counts the number of eigenvalues which have a value less than u, and is referred to as the staircase function. In an interval of length L, $\Delta_3(L)$ is defined as the least square deviation of the staircase function from the best fit to a straight line,

$$\Delta_3(L) = \frac{1}{L}\left\langle \min_{A,B} \int_{u_s}^{u_s+L} (\hat{\eta}(u) - Au - B)^2 du \right\rangle. \tag{5.7}$$

where $\langle\cdot\rangle$ denotes averaging over starting points u_s. When the spectra occurs quite regularly there is a small root-mean-square deviation from a line, and a small contribution to $\Delta_3(L)$. When some of the levels are nearly degenerate, the succession of the stairs becomes irregular, which gives a larger contribution to $\Delta_3(L)$. For the Poisson spectrum without correlations $\Delta_3(L) = L/15$, and for WD spectrum $\Delta_3 = (\ln(2\pi L) + \gamma - 5/4 - \pi^2/8)/\pi^2$, to order $1/L$ [67].

In Figure 5.1 and Figure 5.2, we display the RMT statistics $P(s)$, Σ^2, and Δ_3 for 2-d and 3-d RRN with i.i.d. bonds, respectively. They illustrate that these statistics have the transitional behavior of q-GOEs, described in Section 5.1. More specifically, these simulations demonstrate that for $p \ll 1$ the eigenvalues are virtually uncorrelated, and are governed by Poisson-like statistics, and as p increases and the system becomes increasingly connected, the eigenvalues become increasingly correlated, approaching WD statistics. It is important to note that the spectra of these RRN *can not* be determined by universal WD statistics for any $p \in [0, 1]$ since, as we discussed above, WD statistics are independent of the dimension d of the system, which is inconsistent with what is known about these RRN [60, 116, 122]. However, these simulations demonstrate that the spectra are governed by the one parameter universality class of q-GOEs.

By the symmetry property of the eigenvalue density $\rho(\lambda, p) = \rho(1 - \lambda, 1 - p)$, in the *bulk* of the spectrum, these RMT statistics have the following symmetry in the bulk of the spectrum: $P(s, p) = P(s, 1-p)$, $\Sigma^2(p) = \Sigma^2(1-p)$, and $\Delta_3(p) = \Delta_3(1-p)$. It is important to mention that we performed a local unfolding procedure to obtain these figures. As described in [67], for each $\omega \in \Omega$, this procedure uses a polynomial fit $P_k(\lambda, \omega)$ of the staircase function $\eta(\lambda, \omega) = \int_0^{\lambda} \nu(\lambda', \omega)d\lambda'$ involving the counting measure $\nu(\lambda, \omega) = \sum_j \delta(\lambda - \lambda_j(\omega))$, where k is the order of the polynomial. The variable change in (5.5) is then accomplished using

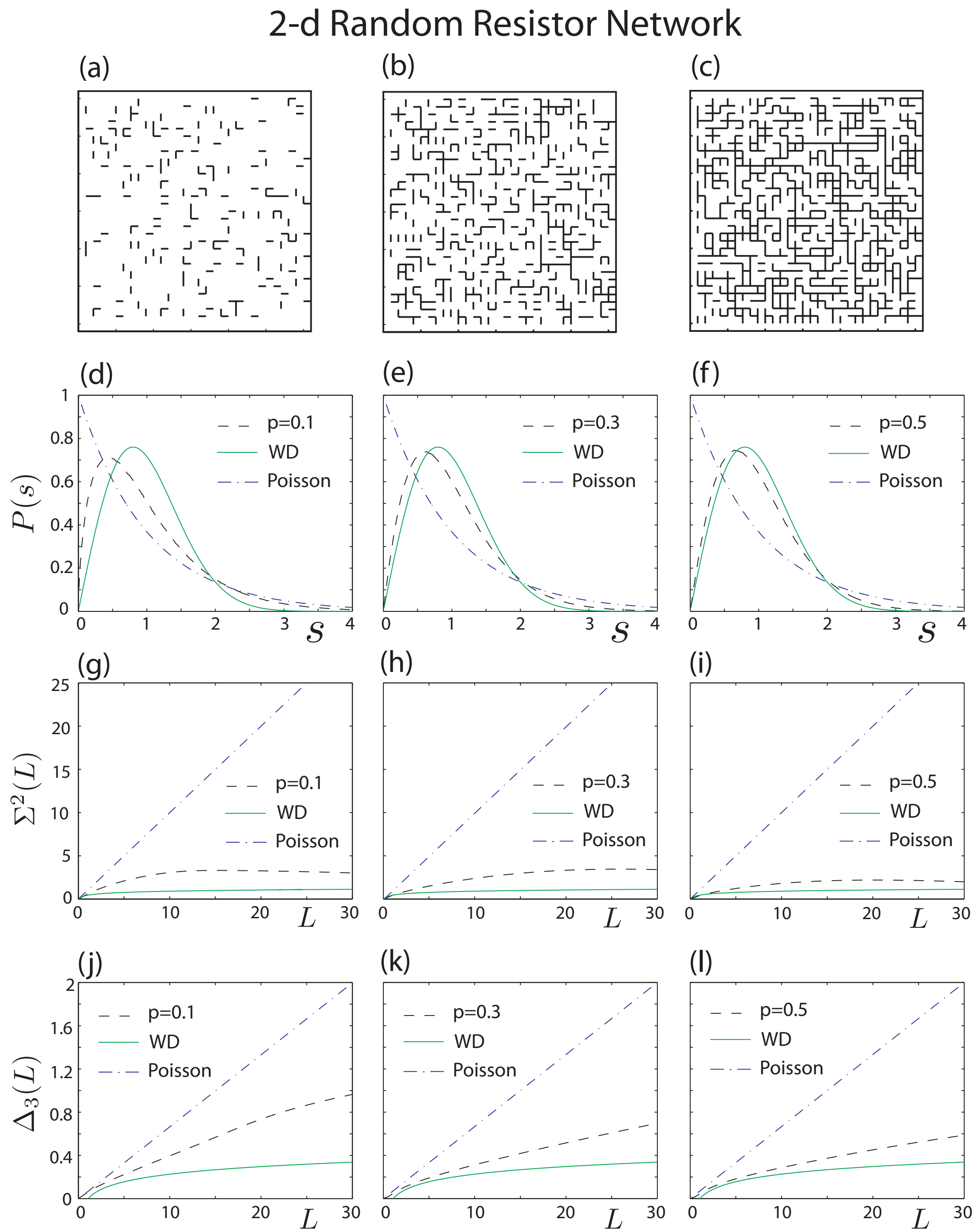

Figure 5.1. Random matrix eigenvalue statistics for 2-d square RRN with i.i.d. bonds. Random realizations of 2-d square RRN are displayed in (a)–(c), with the corresponding eigenvalue statistics (d)–(l) directly below. The conducting regime $p = p_c = 0.5$, is characterized by WD-like statistics which transition, with decreasing p, toward Poisson-like statistics in the insulating regime.

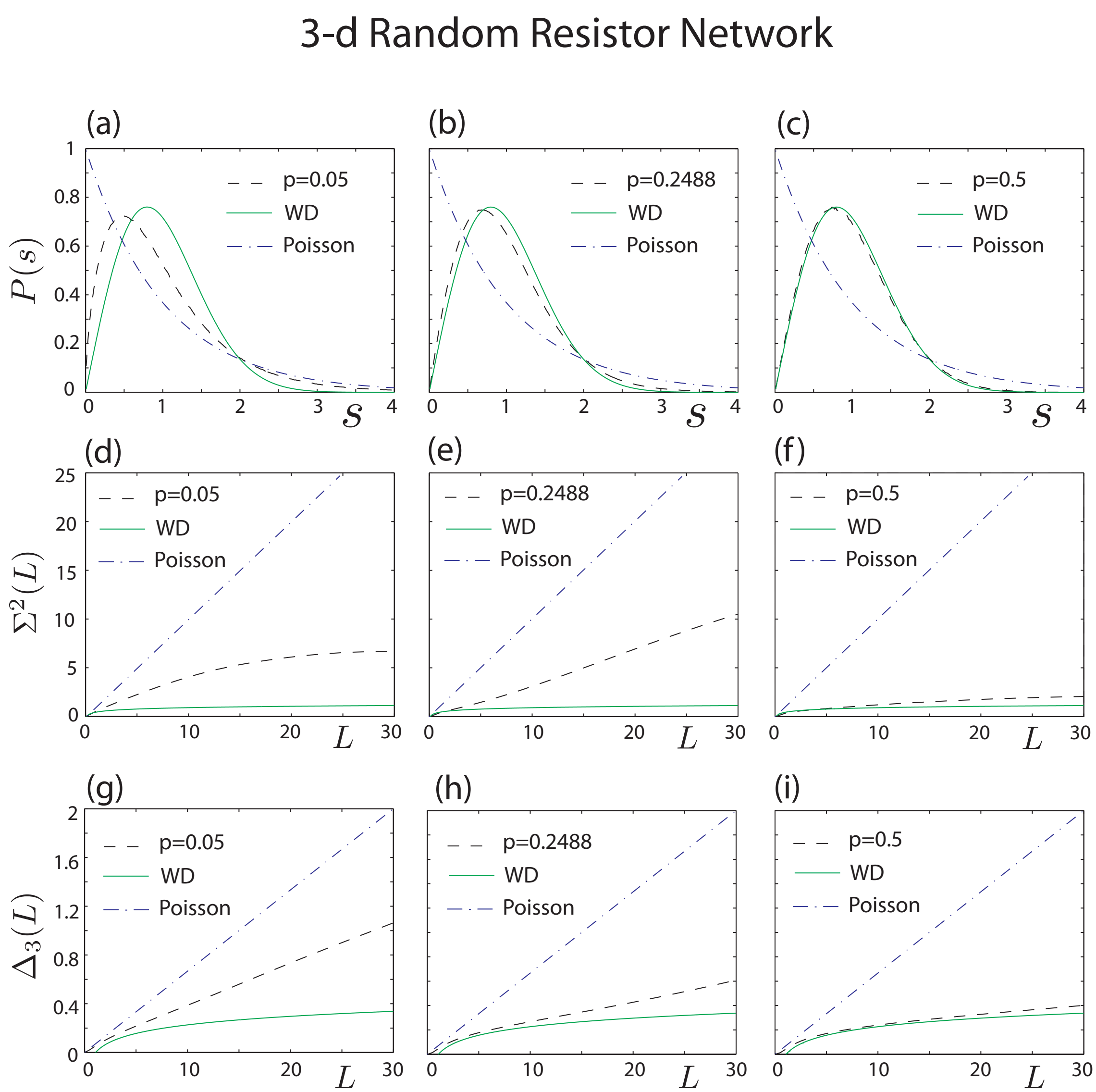

Figure 5.2. Random matrix eigenvalue statistics for 3-d square RRN with i.i.d. bonds. The conducting regime $p \geq p_c \approx 0.2448$, is characterized by WD-like statistics which transition, with decreasing p, toward Poisson-like statistics in the insulating regime.

the local integrated density of states via $u_j = P_k(\lambda_j)$. As can be seen from Figure 5.2 (c), this procedure is quite accurate in the bulk of the spectrum. In order to study eigenvalue correlations near the spectral edges, we need to use the global density ρ evaluated directly from the numerical simulation. However this is highly nontrivial to implement numerically [26, 67], and will be a part of future work. Doing so will also allow us to fit our statistics to that of the mean field theory [26] of the q-GOE, which will provide analytical expressions for these statistics and give deeper insight into critical connectedness transitions exhibited by two-phase random media.

CHAPTER 6

STATISTICAL MECHANICS OF HOMOGENIZATION FOR COMPOSITES

In this chapter we construct, from first principles in physics, a canonical ensemble statistical mechanics framework for general transport models of two-phase dielectric media, and use it study general features of ER fluids. A statistical mechanics model of such a random medium is given entirely by energetic contributions of the system. It is therefore very important to understand these underlying energies. Our novel approach uses an energy representation arising in the ACM, which becomes *exact* in the infinite volume limit. The decomposition of system energy given in Theorem 4 provides detailed information regarding various energetic contributions, and represents them in a way which completely separates parameter information from complicated geometric interactions. Consequently, our physically consistent model is both physically transparent and mathematically tractable.

6.1 Introduction

Thermodynamics was originally a self contained theory of heat and work, based firmly on experimental evidence. Statistical mechanics was subsequently developed to provide a micro-physical foundation for this empirical subject. Thermodynamics was founded as a science by R. Clausius when he gave a kinematic description of thermodynamical systems [9]. He postulated that every thermodynamical system may be characterized by some generalized coordinates (or parameters), which may vary over a physically realizable range. For instance, a solid body experiencing homogeneous deformation may be characterized by the six independent components of the (symmetric) deformation tensor $(e_1, e_2, \cdots, e_6)$ [9, 103]. We will denote generalized coordinates by $y = (y_1, y_2, \ldots, y_n)$. A thermodynamic system is also characterized by the empirical temperature T. The $(n+1)$-dimensional state space of points with coordinates $(y_1, y_2, \ldots, y_n, T)$ describes the thermodynamic state of the system [9].

Clausius formulated two statements, which are now commonly known as the first and second laws of thermodynamics [9]. The first law postulates that every infinitesimal thermodynamic process may be characterized by a work function which is a differential form of the $\{dy_1, dy_2, \ldots, dy_n\}$, $\delta W = \sum_{j=1}^{n} A_i dy_i$, and a heat supply δQ which is a differential form of the $\{dy_1, dy_2, \ldots, dy_n, dT\}$, and that the sum is the differential form of some function of state $U(y_1, y_2, \ldots, y_n, T)$ called the internal energy [9, 18, 103, 121],

$$dU = \delta W + \delta Q = \sum_{j=1}^{n} A_j dy_j + TdS. \tag{6.1}$$

The term TdS is given by the second law, which Clausius derived by arguing: for any cycle, the following equation holds [9]

$$\oint \frac{\delta Q}{T} = 0 \iff \delta Q = TdS. \tag{6.2}$$

The existence of the state function $S(y_1, y_2, \ldots, y_n)$, which Clausius called the entropy, is equivalent to the vanishing of the integral in equation (6.2) [9]. He called T the absolute temperature. Neither δQ nor δW are exact differentials [9, 18, 121]. The symbols δQ and δW are used to indicate the linear differential forms or pfaffins of these functions [18]. Although, dU as well as dS are exact [18]. More generally, the existence of the state functions $T \geq 0$ and S, related by $\delta Q = TdS$, may be established by means of a theorem on canonical presentation of differential forms ([9] and references therein).

By the first and second laws of thermodynamics (6.1) and (6.2), we have

$$\frac{1}{T} = \frac{\partial S(U, y)}{\partial U}, \quad A_j = -T \, \frac{\partial S(U, y)}{\partial y_j}, \quad j = 1, 2, \ldots, n. \tag{6.3}$$

The two laws of thermodynamics are thus reduced to the statement that there exists and entropy function $S(U, y)$ such that the absolute temperature T and generalized forces $\{A_j\}_{j=1}^{n}$ are expressed in terms of the entropy by the constitutive equations (6.3) [9]. The term "entropy" has since been generalized and used in science in many different senses, and has been used in many areas of mathematics and physics, including: dynamical systems, random matrix theory, topology, and information theory [2, 83, 111]. The ideas of information theory and information entropy form a solid foundation for statistical thermophysics [103].

6.1.1 Information Theory and the Canonical Ensemble

In statistical physics, one is faced with the task of assigning probabilities to events associated with complex many body systems, based on a few significant bits of information. In practice, this information is far from sufficient to obtain objective nor unique probabilities. In order to develop a theory that describes macroscopic properties of a system, based on underlying microscopic properties which are not precisely known, it is common to use a maximum entropy principle. The prediction of macroscopic behavior based on insufficient or incomplete data is part of information theory.

An entropy function S is a measure of the amount of uncertainty in a statistical model [103]. The idea behind entropy is that one is not entitled to assume more knowledge, less uncertainty, than that given by subsidiary conditions such as average values and unity measure of the probability space. Any assignment of probabilities that satisfy these conditions but yield a value of S other than its maximum is unjustified on the basis of known data. Therefore, the common attitude is to use probability measures which maximize entropy, thereby maximizing the uncertainty of a system, subject to known information.

The analysis of Shannon [1948] provides a remarkably clear, quantitative measure of the uncertainty inherent in a set of probabilities $\{f_\omega\}$ [6, 103]. He derived the following expression widely known as Shannon's *information entropy*

$$S[\{f_\omega\}] = -k \sum_\omega f_\omega \ln f_\omega, \tag{6.4}$$

where f_ω is the probability of event $\omega \in \Omega$, where Ω is the set of all such events, and k is an arbitrary positive constant which sets the units of S. One can show that the entropy is a strictly concave function [111] with a global minimum $S = 0$, attained when $f(\omega) = 1$ for some $\omega \in \Omega$ (no uncertainty) [103], and with a global maximum $S = k \ln |\Omega|$ attained when $f(\omega) = f(\omega') = 1/|\Omega|$ for all $\omega, \omega' \in \Omega$ [100, 111] (no information), where $|\Omega|$ is the cardinality of the set Ω. Therefore the entropy is inherently positive $S \geq 0$.

A common method for maximizing functions with given constraints is the method of Lagrange multipliers. Of course one always has the constraint $\sum_\omega f_\omega = 1$. When only the average of some quantity is known, $U = \langle U_\omega \rangle = \sum_\omega f_\omega U_\omega$, the resultant probability distribution is called the *canonical ensemble* [103]. The canonical ensemble is found by maximizing $S/k - \alpha \sum_\omega f_\omega - \beta \sum_\omega f_\omega U_\omega$ over probability distributions $\{f_\omega\}$, where α and β are Lagrange multipliers. Regarding the $\{f_\omega\}$ as independent variables, one arrives at the following probability distribution [6, 100, 103]

$$f_\omega = Z^{-1} \exp\left(-\beta U_\omega\right), \quad Z = \sum_\omega \exp(-\beta U_\omega). \tag{6.5}$$

The distribution $\{f_\omega\}$ and its normalization $Z = \exp(\alpha + 1)$ [103] are widely known as the Gibbs–Boltzmann (GB) distribution and the partition function respectively. The exponential nature of the canonical ensemble allows averages to be calculated via the *pressure function* $\ln Z = \alpha + 1$. For example [27, 103],

$$U = \langle U_\omega \rangle = -\frac{\partial \ln Z}{\partial \beta}, \quad \mathrm{Var}(U_\omega) = \langle U_\omega^2 \rangle - \langle U_\omega \rangle^2 = \frac{\partial^2 \ln Z}{\partial \beta^2}, \tag{6.6}$$

where $\langle \cdot \rangle$ denotes averaging with respect to the distribution $\{f_\omega\}$ in (6.5).

The function $\mathcal{F} = -\beta^{-1} \ln Z$ is widely known as the Helmholtz free energy, or simply free energy. Using the free energy, one may write the entropy in the following form [103]

$$U = ST + \mathcal{F}, \quad T = (k\beta)^{-1}, \tag{6.7}$$

where, as we will see in equation (6.8) below, the function U is identified with the internal energy (6.1). Equation (6.7) shows that the internal energy and free energy are Legendre transformations of each other (see Section 6.1.2). In statistical thermophysics the absolute temperature is defined by $T = (k\beta)^{-1}$ [121]. Under the information theoretic approach to statistical mechanics the (universal [100]) constant k is arbitrary. If one sets $k = 1$, then T has units of energy and β is the inverse temperature. If one sets k to Boltzmann's constant, then T has units of Kelvin. In statistical thermophysics, the quantity Q defines processes such as heat transfer and/or radiation [18]. Although, in the information theoretic framework, Q encompasses all energetic processes in which information, $-S$, is lost.

The $\{U_\omega\}$ have been identified by energy states of Hamiltonian systems and the $\{f_\omega\}$ as the corresponding equilibrium distribution [103]. This identification has been generalized to Hamiltonian systems with a continuum of energy states. The macroscopic energy is given by the system Hamiltonian $\mathcal{H}$ and the corresponding partition function is given by $Z = \int_{\omega \in \Omega} P(d\omega) \exp(-\beta \mathcal{H}(\omega))$, where $\omega \in \Omega$ is the space of all statistical realizations, $P(d\omega)$ is the reference measure of the system when $\beta = 0$, and $Z^{-1} P(d\omega) \exp(-\beta \mathcal{H}(\omega))$ is the equilibrium probability (Gibbs) measure. To simplify notation, we will continue to use that of a discrete probability space as its generalization is now clear.

The relation (6.7) was obtained without making any assumptions regarding the nature of the system and is therefore a fundamental relation of the information theoretic approach to statistical mechanics. It is a statement of conservation of energy and is therefore a constraint imposed on the system [18]. In order to see this we look at the differential form of this equation [103]

$$dU = TdS - \sum_\omega f_\omega dU_\omega = \delta Q + \delta W, \tag{6.8}$$

which recovers the first law of Thermodynamics (6.1) and identifies $U = \langle U_\omega \rangle$ with the internal energy. The term $\delta W = -\sum_\omega f_\omega dU_\omega$ represents the differential of work done by the surroundings on the system, changing the characteristic energy states $\{U_\omega\}$ [103]. Conservation of energy in Hamiltonian systems then identifies $-\delta W$ with the work done by the system on the surroundings [4, 121]. Various work terms $A_j dy_j$ may be identified by expanding the differential dU_ω in state variables $\{y_j\}_{j=1}^n$ and examining the physical relevance of the generalized forces $A_j = \langle \partial U_\omega / \partial y_j \rangle$ [103]:

$$dU = TdS - \sum_{j=1}^{n} A_j dy_j, \quad A_j = \left\langle \frac{\partial U_\omega}{\partial y_j} \right\rangle. \tag{6.9}$$

Equations (6.8) and (6.9) recover the constitutive equations in (6.3).

Equation (6.9) gives the differential dU of the internal energy in terms of the state functions T and $\{A_j\}_{j=1}^n$, and state variables S and $\{y_j\}_{j=1}^n$: $U = U(S, y)$, $T = T(S, y)$, and $A_j = A_j(S, y)$. A simple calculation, using equations (6.7) and (6.9), shows that the differential $d\mathcal{F}$ of the free energy is given in terms of the state functions S and $\{A_j\}_{j=1}^n$, and state variables T and $\{y_j\}_{j=1}^n$: $\mathcal{F} = \mathcal{F}(T, y)$, $S = S(T, y)$, $A_j = A_j(T, y)$ [103]

$$d\mathcal{F} = -SdT - \sum_{j=1}^{n} A_j dy_j, \quad S = -\frac{\partial \mathcal{F}}{\partial T} \quad A_j = -\frac{\partial \mathcal{F}}{\partial y_j}. \tag{6.10}$$

The details regarding such Legendre transformations are postponed until section 6.1.2.

In the derivation of the first law (6.8) no assumptions were made about the nature of the system nor the evolution to equilibrium. Therefore, it is valid for reversible, irreversible, quasi-static, and even non-quasi–static evolutions during which the thermodynamic state cannot be defined at all [18]. This is important when one is studying systems with electromagnetic processes which are generally irreversible [18]. See [18, 103, 121] for a detailed discussion of thermodynamic state, reversibility, and related concepts.

If other subsidiary conditions are known, say the averages $\langle g_n \rangle = c_n$ of functions $g_n(U_\omega)$ (again one always has the constraint $f_0 = 1$, $c_0 = 1$), the GB distribution becomes [6, 103]

$$f_\omega = Z^{-1} \exp\left(-\sum_n \beta_n g_n(U_\omega) \right), \quad Z = \sum_\omega \exp\left(-\sum_n \beta_n g_n(U_\omega) \right), \tag{6.11}$$

and the resultant value of S is either reduced or left unchanged [103]. From equation (6.11), one has the analogue of equation (6.7) $S/k = \ln Z + \sum_n \beta_n \langle g_n \rangle$ showing that the entropy and the pressure function are Legendre transforms of one another [103]. If we regard S as a function of the $\{\langle g_n \rangle\}$, we have the generalized constitutive equations

$$\beta_n = \frac{\partial (S/k)}{\partial \langle g_n \rangle}, \quad \langle g_n \rangle = -\frac{\partial \ln Z}{\partial \beta_n} \tag{6.12}$$

giving the $\{\beta_n\}$ in terms of the $\{c_n\}$ [103]. The Helmholtz free energy $\mathcal{F}$ contains the same amount of information as the entropy, and every result which can be calculated from one, can be calculated from the other [103]. Moreover, the symmetric matrices

$$[B_1]_{n,m} = \frac{\partial^2 (S/k)}{\partial \langle g_n \rangle \partial \langle g_m \rangle}, \quad [B_2]_{l,j} = -\frac{\partial^2 \ln Z}{\partial \beta_l \partial \beta_j}, \quad B_1 = B_2^{-1}, \tag{6.13}$$

are inverses of one another [103].

In the information theoretic framework, the second law is a *maximum entropy principle*. It states that the entropy S will increase to a maximum value at equilibrium for isolated systems (fixed total energy and mass), with fixed internal energy U and external state variables (volume, electric field, etc.) [18, 103, 121]. Unconstrained state variables, such as temperature, evolve to the equilibrium values as the entropy becomes maximum [25]. The *minimum internal energy principle* is essentially a restatement of the second law [25]. It states that the internal energy U will decrease to a minimum value at equilibrium for closed systems (only energetic transfers), with fixed entropy S and external state variables.

Mathematically, the second law states that if y_i is an unconstrained variable of state which varies as a system approaches equilibrium, then at equilibrium

$$\left.\frac{\partial S}{\partial y_i}\right|_U = 0, \quad \left.\frac{\partial^2 S}{\partial y_i^2}\right|_U < 0. \tag{6.14}$$

Although, from the properties of an exact differential [103], Legendre transformations [18], and the first law (6.8), we have at equilibrium

$$\left.\frac{\partial U}{\partial y_i}\right|_S = -T \left.\frac{\partial S}{\partial y_i}\right|_U = 0, \quad \left.\frac{\partial^2 U}{\partial y_i^2}\right|_S = -T \left.\frac{\partial^2 S}{\partial y_i^2}\right|_U > 0, \tag{6.15}$$

showing that the internal energy is in fact at a minimum. Therefore, we have shown that (see [25] chapter 5)

$$U_0(S_0) = \inf_{\tilde{y}} U(S_0, \tilde{y}) \tag{6.16}$$

where the minimization is with respect to the unconstrained variables $\tilde{y}$. As the system approaches equilibrium, the unconstrained variables take their equilibrium values and the internal energy U_0 is a function only of the entropy S_0 [25].

Using the maximum entropy principle and the equivalent minimum internal energy principle, one may argue the *minimum Helmholtz free energy principle.* It states that, for closed systems with fixed external state variables and temperature, the Helmholtz free energy is minimized at equilibrium with respect to *any unconstrained internal variables.* To see this let $\tilde{y}$ be the set of unconstrained internal variables. By definition of the Helmholtz free energy and the maximum entropy principle, at equilibrium we have $\mathcal{F}(T,\tilde{y}) = \sup_S(U(S,\tilde{y}) - TS)$. Since $T = (\partial U/\partial S)_{\tilde{y}}$, the maximum occurs when the variable T becomes the equilibrium temperature T_0 [25]. By the minimum internal energy principle (6.16), at equilibrium the Helmholtz free energy will be

$$\begin{aligned}\mathcal{F}_0(T_0) = \sup_{S_0}(U_0(S_0) - T_0S_0) &= \sup_{S_0}(\inf_{\tilde{y}}(U(S_0,\tilde{y})) - T_0S_0) \\ &= \inf_{\tilde{y}}(\sup_{S_0}(U(S_0,\tilde{y}) - T_0S_0)) = \inf_{\tilde{y}}(\mathcal{F}_0(T_0,\tilde{y})),\end{aligned} \tag{6.17}$$

where we have assumed that the order of the extrema can be exchanged. Equation (6.17) states that the Helmholtz free energy is minimized at equilibrium. These physical arguments [25] can be made rigorous using concepts of measure theory, free entropy, specifications, and large deviation theory under Gibbs measures [100].

6.1.2 Thermodynamic Potentials and Maxwell's Relations

In order to use the methods of statistical mechanics to uniquely determine the macroscopic behaviors of a system, one must assume a set of constitutive relations which define the state variables and the state functions [18]. It is typically assumed that the functions of state are invertible, at least locally. Therefore, the functions of state and state variables may change roles.

The change of variables is accomplished through Legendre transformations [4, 18, 103]. Through these transformations, various thermodynamic potentials determine the corresponding state functions through constitutive equations like those in equations (6.9) and (6.10). Depending on which variables are chosen as state variables, one may use different thermodynamic potentials which make calculations of certain functions of state much easier [25] as, in the new coordinate system, the state functions are simply derivatives of the corresponding thermodynamic potential with respect to the conjugate state variables.

For a concrete example, we now derive equation (6.10). By equations (6.7)-(6.9), the internal energy $U = U(S, y)$ is a function of the entropy S and the state variables $y = (y_1, \ldots, y_n)$, with total differential $dU = TdS - \sum_{j=1}^{n} A_j dy_j$. We claim that the free energy $\mathcal{F}$, given by the Legendre transformation $\mathcal{F} = U - TS$ in (6.7), is a function of the temperature T and the state variables $y = (y_1, \ldots, y_n)$, $\mathcal{F} = \mathcal{F}(T, y)$, with total differential $d\mathcal{F} = -SdT - \sum_{j=1}^{n} A_j dy_j$. Indeed,

$$d\mathcal{F} = dU - TdS - SdT = TdS - \sum_{j=1}^{n} A_j dy_j - TdS - SdT = -SdT - \sum_{j=1}^{n} A_j dy_j.$$

In this way, we may define many "thermodynamic potentials" U, $\mathcal{F}$, etc.

Often a thermodynamic system is completely described by the absolute temperature T and two state variables y_1 and y_2 with conjugate state functions A_1 and A_2, say. For example, consider a vessel filled with a gas, separated by a semi-permeable membrane, and one of the walls is a movable piston [103, 121]. In this case, $y_1 = V$ is the volume of the vessel, $A_1 = P$ is the external pressure on the piston, $y_2 = N$ is the number of gas molecules to the left of the membrane, say, and $A_2 = \mu$ is the chemical potential.

In Section 1.2 and Section 2.1, we found that the natural state variables for two-phase dielectric media are given by: the average electric field strength $E_0 = y_1$ and the dielectric contrast parameter $t = y_2$, where $t = 1/(1 - \epsilon_2/\epsilon_1)$. The natural conjugate state functions are the effective dipole moment $\mathrm{p}^* = A_1$ and the "contrast potential" $\Psi = A_2$. As this is the system of interest in this chapter, we will henceforth use this notation.

We will see in Section 6.2, that the parameter separation property of the system energy, displayed in equations (2.4), (2.5), and (2.13) of Section 2.1, yields an especially convenient representation of p^*, involving the Stieltjes transform $G(t; \alpha)$ of the spectral measure α introduced in Section 2.1. From this representation and the Maxwell's relations given in equations (6.18)-(6.20) below, we will show that the contrast potential Ψ and electric component of the entropy S are explicitly given by GB canonical ensemble averages of Herglotz functions involving α. Moreover, the decomposition of energy given in Theorem 4 then explicitly identifies the contrast potential Ψ with a specific component of the system energy. This in turn, leads to a physically consistent statistical mechanics model of two-phase dielectric media which is both physically consistent and mathematically tractable.

Maxwell's relations are found by equating commuted mixed partial derivatives of thermodynamic potentials, with respect to the state variables. The following formulas summarize Maxwell's relations for two-phase dielectric media.

$$\mathcal{F} = U - TS, \qquad d\mathcal{F} = -SdT - \mathrm{p}^* dE_0 - \Psi dt \tag{6.18}$$
$$-\frac{\partial^2 \mathcal{F}}{\partial E_0 \partial T} = \frac{\partial S}{\partial E_0} = \frac{\partial \mathrm{p}^*}{\partial T}$$
$$-\frac{\partial^2 \mathcal{F}}{\partial E_0 \partial t} = \frac{\partial \Psi}{\partial E_0} = \frac{\partial \mathrm{p}^*}{\partial t}$$
$$-\frac{\partial^2 \mathcal{F}}{\partial T \partial t} = \frac{\partial S}{\partial t} = \frac{\partial \Psi}{\partial T}$$

$$\mathcal{G} = \mathcal{F} + \mathrm{p}^* E_0, \qquad d\mathcal{G} = -SdT + E_0 d\mathrm{p}^* - \Psi dt \tag{6.19}$$
$$-\frac{\partial^2 \mathcal{G}}{\partial t \partial \mathrm{p}^*} = -\frac{\partial E_0}{\partial t} = \frac{\partial \Psi}{\partial \mathrm{p}^*}$$
$$-\frac{\partial^2 \mathcal{G}}{\partial T \partial \mathrm{p}^*} = -\frac{\partial E_0}{\partial T} = \frac{\partial S}{\partial \mathrm{p}^*}$$

$$\Phi = \mathcal{F} + t\Psi, \qquad d\Phi = -SdT - \mathrm{p}^* dE_0 + t d\Psi \tag{6.20}$$
$$-\frac{\partial^2 \Phi}{\partial T \partial \Psi} = -\frac{\partial t}{\partial T} = \frac{\partial S}{\partial \Psi},$$

where $\mathcal{F}$ is the Helmholtz free energy, $\mathcal{G}$ is the Gibbs free energy, and Φ is the Grand potential [103]. For this system, there are a total of eight thermodynamic potentials, with three Maxwell's relations each. However the identity [103] $\partial a/\partial b = (\partial b/\partial a)^{-1}$ makes many of these relations redundant. It is true that any of these statistical mechanics potentials determines any other. For example [4]

$$U = \mathcal{F} + TS = \mathcal{F} - T\frac{\partial \mathcal{F}}{\partial T} \tag{6.21}$$
$$\mathcal{G} = \mathcal{F} + \mathrm{p}^* E_0 = \mathcal{F} - E_0 \frac{\partial \mathcal{F}}{\partial E_0}$$
$$\mathcal{F} = \mathcal{G} - \mathrm{p}^* E_0 = \mathcal{G} - \mathrm{p}^* \frac{\partial \mathcal{G}}{\partial \mathrm{p}^*}.$$

However, the formulas in equation (6.21) indicate why certain thermodynamic potentials make calculations of certain state functions much more convenient.

We will see in section 6.2 that Maxwell's relations provide important information regarding phase transitions exhibited by two-phase dielectric media. These results are physically consistent with experimental observations of ER fluids [126]. In section 6.1.3 we first review the statistical origin of the temperature T arising in the canonical ensemble of statistical mechanics.

6.1.3 Temperature and the Generalized Equipartition Theorem

Within the canonical ensemble, the variables that naturally characterize the macroscopic state of the system are the temperature T, state variables which describe intrinsic properties of the system such as the particle number N or the dielectric contrast parameter t, and state variables which describe external work done on the system such as the volume V or the average electric field strength E_0. Energy fluctuations are allowed since the system has been placed in direct thermal contact with an external heat bath, or temperature reservoir, with fixed temperature T. Consequently, the temperature naturally appears in various canonical ensemble averages, whereby one can obtain useful relations between various mechanical quantities and T.

The mathematical meaning of the temperature, within the canonical ensemble, is given by the *Generalized Equipartition Theorem* [123]. Consider a matrix/particle system, and denote by $\vec{p_i} = (p_1, \ldots, p_d)_i$ and $\vec{r_i} = (r_1, \ldots, r_d)_i$ the linear momentum and position vectors of particle $i = 1, \ldots, N$, respectively, where d is the dimension of the system. The partition function Z is given by [123]

$$Z = \int_{\mathbb{R}^{2dN}} e^{-\beta\mathcal{H}} d\vec{p}_1 \cdots d\vec{p}_N d\vec{r}_1 \cdots d\vec{r}_N, \tag{6.22}$$

where $\beta = 1/kT$, k is Boltzmann's constant, and $\mathcal{H}$ is the Hamiltonian of the system. We assume that the Hamiltonian has the following asymptotic behavior

$$\lim_{|\vec{p}_i| \longrightarrow \infty} \exp(-\beta\mathcal{H}) = \lim_{|\vec{r}_i| \longrightarrow \infty} \exp(-\beta\mathcal{H}) = 0, \tag{6.23}$$

for all $i = 1, \ldots, N$. Integration by parts then yields the *Equipartition Theorem* [123]:

$$\left\langle p_m \frac{\partial\mathcal{H}}{\partial p_n} \right\rangle = Z^{-1} \int_{\mathbb{R}^{2dN}} p_m(-kT) \frac{\partial e^{-\beta\mathcal{H}}}{\partial p_n} dp_1 \cdots dp_{dN} dr_1 \cdots dr_{dN} = kT\delta_{nm}, \tag{6.24}$$

where δ_{nm} is the Kronecker delta. Similarly, using r_i in lieu of the p_i, we obtain the *Virial Theorem*, $\langle r_m \partial\mathcal{H}/\partial r_n \rangle = kT\delta_{nm}$. Together we have

$$\langle \vec{p}_m \cdot \nabla_{\vec{p}_n} \mathcal{H} \rangle = \langle \vec{r}_m \cdot \nabla_{\vec{r}_n} \mathcal{H} \rangle = dkT\delta_{nm}. \tag{6.25}$$

Equation (6.25) is called the Generalized Equipartition Theorem. It indicates that each particle is coupled to the external bath in an identical manner. When derived within the canonical ensemble, the above relations are valid for any system size (neglecting the energy of interaction between particles within the system and particles within the heat bath) [123]. Moreover, the explicit form of the Hamiltonian is not needed, and the theorem is valid for any Hamiltonian system satisfying (6.23).

Consider a more explicit form of the system Hamiltonian:

$$\mathcal{H} = \sum_{j=1}^{N} \frac{|\vec{p}_j|^2}{2m_j} + I(\vec{r}_1, \ldots, \vec{r}_N), \tag{6.26}$$

where $|\vec{p}_j|^2/2m_j$ is the kinetic energy particle j of mass m_j and I is the inter-particle interaction potential satisfying $\lim_{|\vec{r}_j| \longrightarrow \infty} I = \infty$ for all $j = 1, \ldots, N$. In this case we obtain a well known consequence of the Equipartition Theorem [123]:

$$\left\langle \frac{\vec{p}_i \cdot \vec{p}_j}{2m_j} \right\rangle = \frac{dkT}{2}\delta_{ij}, \quad \Rightarrow \quad \sum_{j=1}^{N} \left\langle \frac{|\vec{p}_j|^2}{2m_j} \right\rangle = \frac{dNkT}{2}, \tag{6.27}$$

which follows by direct substitution of formula (6.26) in equation (6.25). Equation (6.27) states that the average kinetic energy of each degree of freedom is given by $dkT/2$, i.e., the kinetic energy is equally partitioned between its degrees of freedom. This secondary result is known as the *Principle of Equipartition*. It is important to note that the Generalized Equipartition Theorem (6.25) is the fundamental result from which one determines how the translational degrees of freedom are related to the temperature of the heat bath. The equal sharing of kinetic energy is a secondary result that follows from taking the kinetic energy to be a homogeneous quadratic function of the generalized momenta, and is not a necessary consequence of Hamiltonian systems. In fact, the Generalized Equipartition Theorem (6.25) holds in systems where the principle of equipartition (6.27) is no longer valid [123].

In section 6.1.4 we explore general, asymptotic features of canonical ensemble statistical mechanics models, of Hamiltonian systems with a finite number of energy levels. There we show that for high temperatures $T \gg 1$, the system asymptotically behaves according to the reference measure. While for $T \ll 1$, the system evolves toward configurations of low intrinsic energy, due to an exponentially decreasing probability of high intrinsic energy as the temperature decreases.

6.1.4 Asymptotic Analysis of the Canonical Ensemble

In this section, we explore how phase transitions in the canonical ensemble may be characterized by transitions in the GB probability measure, its energy moments, Helmholtz free energy, and entropy. We show that for high temperatures, $T \gg 1$ ($\beta \ll 1$), the system behaves according to the reference probability measure, and as the temperature vanishes, $T \longrightarrow 0$ ($\beta \longrightarrow \infty$), the probability measure becomes highly localized about the system's minimum energy state. In the canonical ensemble, a phase transition is characterized by a sudden change in the behavior of the probability measure, largely deviating away from the reference measure (RM), near a critical temperature T_c, towards the delta measure concentrated at the minimum energy state. This asymptotic analysis will provide us with intuition for the general transport models of two-phase dielectric media discussed in section 6.2.

For simplicity, consider a Hamiltonian system with a finite number of (distinct) energy states

$$-\infty < \mathcal{H}_{\min} = \mathcal{H}_1 < \mathcal{H}_2 < \cdots < \mathcal{H}_M = \mathcal{H}_{\max} < \infty. \tag{6.28}$$

The above assumptions allow us to put the underlying probability space Ω into one-to-one correspondence with the set of integers $\Omega = \{1, 2, \ldots, M\}$, with $|\Omega| = M$. Therefore, the RM is the uniform distribution over this set. In general, the RM can be much more complicated. For example, in ER fluids, the underlying RM $P(d\omega)$ characterizes the modified Poisson distribution of hard noninteracting spheres. Theorem 10 is the main result of this section.

Theorem 10 *Let $\mathcal{H}_{min}$ and $\mathcal{H}_{max}$ be defined as in equation* (6.28)*, and denote by $\mathcal{H}_\omega$ the energy of state $\omega \in \Omega$, $\Delta\mathcal{H}_\omega = \mathcal{H}_\omega - \mathcal{H}_{min}$, $f_\omega = Z^{-1}\exp(-\beta\mathcal{H}_\omega)$ the GB probability distribution with average $\langle\cdot\rangle_{\mathcal{H}}$ and variance $Var(\cdot)_{\mathcal{H}}$, $Z = \sum_\omega \exp(-\beta\mathcal{H}_\omega)$ its normalizing partition function, β the inverse temperature, $\mathcal{F} = -\beta^{-1}\ln Z$ the free energy, $S = (\langle\mathcal{H}_\omega\rangle_{\mathcal{H}} - \mathcal{F})/T$ the entropy, $\langle\cdot\rangle$ averaging with respect to the RM, and $Var(\cdot)$ variance with respect to the RM. Then in the high temperature regime, $\beta \ll 1$, we have*

(1) $f_\omega = |\Omega|^{-1} + O(\beta)$, (2) $\langle\mathcal{H}_\omega^n\rangle_{\mathcal{H}} = \langle\mathcal{H}_\omega^n\rangle + O(\beta)$,

(3) $Var(\mathcal{H}_\omega)_{\mathcal{H}} = Var(\mathcal{H}_\omega) + O(\beta)$ (4) $\mathcal{F} = -\beta^{-1}\ln|\Omega| + \langle\mathcal{H}_\omega\rangle + O(\beta)$,

(5) $S = k\ln|\Omega| + O(\beta/T)$

Moreover, for $\beta \gg 1$ the low temperature regime

(6) $f_\omega = e^{-\beta\Delta\mathcal{H}_\omega} + O(\exp(-\beta\Delta\mathcal{H}_2))$, (7) $\langle\mathcal{H}_\omega^n\rangle_{\mathcal{H}} = \mathcal{H}_{min}^n + O(\exp(-\beta\Delta\mathcal{H}_2))$,

(8) $Var(\mathcal{H}_\omega)_{\mathcal{H}} = O(\exp(-\beta\Delta\mathcal{H}_2))$ (9) $\mathcal{F} = \mathcal{H}_{min} + O(\beta^{-1}\exp(-\beta\Delta\mathcal{H}_2))$,

(10) $S = O(\beta\exp(-\beta\Delta\mathcal{H}_2))$

Before we prove Theorem 10, we state what is says. For $\beta \ll 1$, to $O(\beta)$, it says that the probability measure is given by the RM, the energy moments and the variance are given by that with respect to the RM , the free energy $\mathcal{F}$ is given by that of the RM plus the average energy with respect to the RM, and to $O(\beta/T)$ the entropy S is given by its global maximum. For $\beta \gg 1$, to $O(\exp(-\beta\Delta\mathcal{H}_2))$, the probability measure behaves like the delta measure concentrated at the minimum energy $\mathcal{H}_{\min}$, the nth energy moment is the nth power of $\mathcal{H}_{\min}$, the variance is exponentially small, to $O(\beta^{-1}\exp(-\beta\Delta\mathcal{H}_2))$ the free energy $\mathcal{F}$ is given by $\mathcal{H}_{\min}$ and the entropy is exponentially small.

Proof: First, when $\beta \ll 1$, we may expand $\exp(-\beta\mathcal{H}_\omega)$ in a Taylor series, and the partition function may be represented by

$$Z = \sum_\omega e^{-\beta\mathcal{H}_\omega} = |\Omega|(1 + V_0(\{\mathcal{H}_\omega\})), \quad V_j(\{\mathcal{H}_\omega\}) = \langle\tilde{V}_j\rangle = \sum_{n=1}^{\infty} \frac{(-1)^n\beta^n}{n!}\langle\mathcal{H}_\omega^{n+j}\rangle, \tag{6.29}$$

where $\langle\cdot\rangle$ denotes averaging with respect to the RM. Therefore by the geometric series, the GB probability measure is given by

$$f_\omega = |\Omega|^{-1}(1+\tilde{V}_0)(1+R(\{\mathcal{H}_\omega\})), \quad R(\{\mathcal{H}_\omega\}) = \sum_{j=1}^{\infty}[-V_0(\{\mathcal{H}_\omega\})]^j, \tag{6.30}$$

$$f_\omega \sim |\Omega|^{-1} + \beta(\mathcal{H}_\omega - \langle\mathcal{H}_\omega\rangle) + \frac{\beta^2}{2}[\mathcal{H}_\omega^2 - \langle\mathcal{H}_\omega^2\rangle - 2(\mathcal{H}_\omega\langle\mathcal{H}_\omega\rangle - \langle\mathcal{H}_\omega\rangle^2)] + O(\beta^3),$$

where $\beta \ll 1$ has been chosen so that $|V_0| < 1$. Equation (6.30) shows that, for $\beta \ll 1$, the GB probability measure behaves like the reference probability measure, to $O(\beta)$.

By equation (6.30) and the identity $\tilde{V}_n = \mathcal{H}_\omega^n\tilde{V}_0$, the energy moments $\langle\mathcal{H}_\omega^n\rangle_\mathcal{H}$ of the GB probability measure are given by

$$\begin{aligned}\langle\mathcal{H}_\omega^n\rangle_\mathcal{H} &= [\langle\mathcal{H}_\omega^n\rangle + V_n(\{\mathcal{H}_\omega\})][1 + R(\{\mathcal{H}_\omega\})] \\ &\sim \langle\mathcal{H}_\omega^n\rangle - \beta\mathrm{Cov}(\mathcal{H}_\omega^n, \mathcal{H}_\omega) + \frac{\beta^2}{2}[\mathrm{Cov}(\mathcal{H}_\omega^n, \mathcal{H}_\omega^2) - 2\langle\mathcal{H}_\omega\rangle\mathrm{Cov}(\mathcal{H}_\omega^n, \mathcal{H}_\omega)] + O(\beta^3),\end{aligned}$$

where $\langle\cdot\rangle_\mathcal{H}$ denotes the GB average and $\mathrm{Cov}(\cdot,\cdot)$ is the covariance with respect to the RM. In particular, the GB variance of the Hamiltonian, $\mathrm{Var}(\mathcal{H}_\omega)_\mathcal{H}$, satisfies

$$\mathrm{Var}(\mathcal{H}_\omega)_\mathcal{H} \sim \mathrm{Var}(\mathcal{H}_\omega) - \beta[\mathrm{Cov}(\mathcal{H}_\omega^2, \mathcal{H}_\omega) - 2\langle\mathcal{H}_\omega\rangle\mathrm{Var}(\mathcal{H}_\omega)] + O(\beta^2), \tag{6.31}$$

where $\mathrm{Var}(\cdot)$ is the variance with respect to the RM. By equation (6.29), the Helmholtz free energy $\mathcal{F}$ and the entropy S are given by

$$\begin{aligned}\mathcal{F} &= -\beta^{-1}\ln Z = -\beta^{-1}\ln|\Omega| - \beta^{-1}\ln(1+V_0) \\ &\sim -\beta^{-1}\ln|\Omega| + \langle\mathcal{H}_\omega\rangle - \frac{\beta}{2}\mathrm{Var}(\mathcal{H}_\omega) + O(\beta^2), \\ S &= (\langle\mathcal{H}_\omega\rangle_\mathcal{H} - \mathcal{F})/T \sim k\ln|\Omega| - \frac{\beta}{2T}\mathrm{Var}(\mathcal{H}_\omega) + O\left(\frac{\beta^2}{T}\right).\end{aligned} \tag{6.32}$$

Therefore, to $O(\beta)$, when $\beta \ll 1$ the Helmholtz free energy follows the average energy $\langle\mathcal{H}_\omega\rangle$ and, to $O(\beta/T)$, the entropy is at its global maximum $S = k\ln|\Omega|$.

In the opposite limit, $\beta \gg 1$, the partition function is represented by

$$Z = \sum_{\omega} e^{-\beta \mathcal{H}_\omega} = e^{-\beta \mathcal{H}_{\min}} \left(1 + \sum_{\omega \neq 1} e^{-\beta \Delta \mathcal{H}_\omega} \right) = e^{-\beta \mathcal{H}_{\min}} (1 + Z_\Delta), \tag{6.33}$$

where $\Delta \mathcal{H}_\omega = \mathcal{H}_\omega - \mathcal{H}_{\min} \geq 0$ with equality only if $\omega = 1$. Therefore, by the geometric series, the GB probability measure is given by

$$f_\omega = e^{-\beta \Delta \mathcal{H}_\omega} \left(1 + \sum_{j=1}^{\infty} (-Z_\Delta)^j \right), \tag{6.34}$$

where $\beta \gg 1$ has been chosen so that $Z_\Delta \sim e^{-\beta \Delta \mathcal{H}_2} < 1$. As $\exp(-\beta \Delta \mathcal{H}_\omega) \ll 1$ for all $\omega \neq 1$ when $\beta \gg 1$, Z_Δ is exponentially small. Therefore, equation (6.34) and the identity $\exp(-\beta \Delta \mathcal{H}_1) \equiv 1$ shows that the GB probability measure is highly localized about the system minimum energy $\mathcal{H}_{\min}$.

By equation (6.34), the energy moments $\langle \mathcal{H}_\omega^n \rangle_{\mathcal{H}}$ of the GB probability measure are given by

$$\begin{aligned} \langle \mathcal{H}_\omega^n \rangle_{\mathcal{H}} &= \left(\mathcal{H}_{\min}^n + \sum_{\omega \neq 1} \mathcal{H}_\omega^n e^{-\beta \Delta \mathcal{H}_\omega} \right) \left(1 + \sum_{j=1}^{\infty} (-Z_\Delta)^j \right) \\ &\sim \mathcal{H}_{\min}^n + (\mathcal{H}_2^n - \mathcal{H}_{\min}^n) e^{-\beta \Delta \mathcal{H}_2} + O\left(e^{-\beta \Delta \mathcal{H}_3}\right) \end{aligned} \tag{6.35}$$

In particular, the GB variance $\mathrm{Var}(\mathcal{H}_\omega)_{\mathcal{H}}$ of the Hamiltonian satisfies

$$\mathrm{Var}(\mathcal{H}_\omega)_{\mathcal{H}} \sim \Delta \mathcal{H}_2^2 e^{-\beta \Delta \mathcal{H}_2} + O\left(e^{-\beta \Delta \mathcal{H}_3}\right). \tag{6.36}$$

This indicates that the GB variance of the Hamiltonian is exponentially small when $\beta \gg 1$, and vanishes exponentially fast in the limit $\beta \longrightarrow \infty$, so that the GB probability measure approaches the delta measure concentrated at the minimum energy $\mathcal{H}_{\min}$. By equation (6.33), the Helmholtz free energy $\mathcal{F}$ and the entropy S are given by

$$\begin{aligned} \mathcal{F} &= -\beta^{-1} \ln Z = \mathcal{H}_{\min} - \beta^{-1} \ln(1 + Z_\Delta) = \mathcal{H}_{\min} + O\left(\beta^{-1} e^{-\beta \Delta \mathcal{H}_2}\right) \\ S &= (\langle \mathcal{H}_\omega \rangle_{\mathcal{H}} - \mathcal{F})/T = O\left(\beta e^{-\beta \Delta \mathcal{H}_2}\right). \end{aligned} \tag{6.37}$$

Therefore when $\beta \gg 1$, to exponential order, the Helmholtz free energy follows the minimum energy $\mathcal{H}_{\min}$, and the entropy is at its global minimum $S = 0$.

In conclusion, we gave an asymptotic analysis of the GB probability measure, its energy moments, Helmholtz free energy, and entropy, for arbitrary Hamiltonian systems with a finite number of energy levels. We demonstrated that for $\beta \ll 1$, the system evolves according to the reference probability measure, while for $\beta \gg 1$ the system evolves according to a probability measure which is highly localized about the system minimum energy $\mathcal{H}_{\min}$. We say that the system undergoes a *phase transition* when there is a crossover between these two regimes, near a critical temperature T_c, which is so dramatic that the Helmholtz free energy $\mathcal{F}$ loses its analytic properties (in the infinite volume limit). This is the well known behavior of the Ising model, shown in Figures 2.26 and 2.27 in [36], which describes the phenomena of spontaneous magnetization near the Curie temperature. While the existence of a phase transition for general transport models of two-phase dielectric media is beyond the scope of this dissertation, in Section 6.2 we demonstrate that these general models have a behavior analogous to that shown in this section.

6.2 Statistical Mechanics for Two-Phase Dielectric Media

In this section we present a canonical ensemble statistical mechanics framework for general transport models of two-phase dielectric media. In Section 6.2.1 we derive, from first principles in physics, the system Hamiltonian and electric work term for such media. In Section 6.2.2 we explore the consequences of Maxwell's relations. There, we demonstrate that the electric components of state functions are determined by the GB statistics of Herglotz functions, which arise in the decomposition of system energy given in Theorem 4. The physical consistency, local stability, and asymptotic analysis of our model is given in Theorem 11.

6.2.1 The System Hamiltonian and Electric Work Term

In this section we derive an appropriate system Hamiltonian $\mathcal{H}_\omega$ and electric work term δW for general transport models of two-phase dielectric media. By the linearity of the governing equations [87], the Hamiltonian is the sum of pure electric energies, pure elastic energies, pure thermal energies, and mutual coupling energies [103]. In order to model two-phase dielectric media using methods of statistical mechanics, we must decompose the system Hamiltonian into these various contributions and identify the associated work terms, to be inserted into the first law (6.18) for such media. When multiple energetic sources are considered which exhibit coupling, such as the electric/elastic phenomenon of electrostriction exhibited by all dielectrics, this decomposition can be highly nontrivial [18]. For simplicity, we focus solely on the electric aspects of two-phase dielectric media.

From equation (1.30) of Section 1.2, the electric energy (density) associated with an *infinite* two-phase dielectric medium is given by

$$\frac{1}{2}\langle\vec{D}\cdot\vec{E}\rangle = \frac{1}{2}\epsilon^* E_0^2, \quad \epsilon^* = \epsilon_1(1 - G(t;\alpha)), \tag{6.38}$$

where $\langle\cdot\rangle$ will denote volume averaging in this section. Recall from Section 1.2 that the local electric field $\vec{E}$ is given by $\vec{E} = \vec{E}_0 + \vec{E}_f$, where $\vec{E}_f$ is the fluctuating electric field of mean zero $\langle\vec{E}_f\rangle = 0$ about the (constant) average $\langle\vec{E}\,\rangle = \vec{E}_0 = E_0\vec{e}_0$, the unit vector $\vec{e}_0$ is the direction of $\vec{E}_0$ and E_0 is its magnitude, $\epsilon^* = \boldsymbol{\epsilon}^*\vec{e}_0\cdot\vec{e}_0$, $\boldsymbol{\epsilon}^*$ is the effective complex permittivity tensor, $G(t;\alpha)$ is the Stieltjes transform of α, defined in (2.13), which is a spectral measure of the operator $\Gamma\chi_2$, $t = 1/(1-\epsilon_2/\epsilon_1)$, and $\langle\vec{D}\cdot\vec{E}_f\rangle = 0$.

Here we consider a *finite* two-phase dielectric medium contained in $\mathcal{V}\subset\mathbb{R}^d$ with volume V satisfying $1 \ll V < \infty$. As $\langle\vec{D}\cdot\vec{E}_f\rangle = 0$ and $\langle\vec{E}_f\rangle = 0$ for an infinite system, we have for this finite system the existence of constants $a, b > 0$ such that $\langle\vec{D}\cdot\vec{E}_f\rangle = o(V^{-a})$ and $\langle\vec{E}_f\rangle = o(V^{-b})$ as $V \longrightarrow \infty$ [65]. As the infinite volume, thermodynamic limit of our statistical mechanics model is beyond the scope of this dissertation, we simplify our notation and streamline our analysis by dropping the $o(V^{-a})$ and $o(V^{-b})$ notation, associated with setting these quantities to zero. For now on we will denote by $\langle\cdot\rangle$ spatial average over the region $\mathcal{V}$.

For this finite system, effective quantities depend on boundary conditions and the geometric configuration $\omega\in\Omega$ through a family of spectral measures $\{\alpha_\omega\}_{\omega\in\Omega}$:

$$\frac{1}{2}\langle\vec{D}\cdot\vec{E}\rangle = \frac{1}{2}\epsilon^*_\omega E_0^2, \quad \epsilon^*_\omega = \epsilon_1(1+\chi^*_\omega(t)), \quad \chi^*_\omega(t) = -G(t;\alpha_\omega), \tag{6.39}$$

where $\chi^*_\omega(t)$ plays the role of the electric susceptibility and Ω denotes the space of all geometric configurations accessible to the dielectric medium, contained in the vessel $\mathcal{V}$ with volume V. These physically accessible configurations are determined by the reference probability measure $P(d\omega)$ defined in Section 2.1. A key feature of equation (6.39) is that parameter information in t and E_0 is *separated* from complicated geometric interactions, which are incorporated in the $\{\alpha_\omega\}_{\omega\in\Omega}$ [65]. For simplicity we assume that t is real with $\epsilon_2 > \epsilon_1$, so that $t < 0$ and $-G(t;\alpha_\omega) > 0$ [89]. For each $\omega\in\Omega$, $G(t;\alpha_\omega)$ is an analytic function of t for all $t < 0$ [65], and is uniformly bounded for all $t < -\varepsilon < 0$ [89]. We will see that these properties are inherited by the system Hamiltonian $\mathcal{H}_\omega$, and yield an especially convenient representation for the electric work term δW.

We now derive the system Hamiltonian and electric work term, following the suggestive analysis of Robertson for homogeneous dielectric media [103]. Since $\vec{E} = \vec{E}_0 + \vec{E}_f$ and $\vec{D} = \epsilon\vec{E}$, the energy is given by

$$\left\langle \frac{1}{2}\epsilon\left(E_0^2 + 2\vec{E}_f \cdot \vec{E}_0 + E_f^2\right)\right\rangle = \mathcal{W}_0 + \mathcal{W}_{int} + \mathcal{W}_f = \mathcal{W}_0 + \frac{1}{2}\mathcal{W}_{int}, \tag{6.40}$$

as $\langle \vec{D} \cdot \vec{E}_f \rangle = \frac{1}{2}\mathcal{W}_{int} + \mathcal{W}_f = 0$. Recalling that $\epsilon = \epsilon_1(1 - \chi_2/t)$ and $\langle \vec{E}_f \rangle = 0$, we have

$$\mathcal{W}_0 + \frac{1}{2}\mathcal{W}_{int} = \frac{1}{2}\epsilon_1 E_0^2 \left(1 - \frac{p}{t} - \left(G(t; \alpha_\omega) - \frac{p}{t}\right)\right), \tag{6.41}$$

where $p = \langle \chi_2 \rangle$ is the volume fraction of material phase 2. Each term in equation (6.40) must be analyzed in order to correctly obtain the Hamiltonian and electric work term, to be inserted in the first law of thermodynamics (6.18) for binary dielectrics.

By the linearity of Maxwell's equations, the Hamiltonian is the sum of Coulomb energy terms representing the potential energy of all charged particles associated with the system [103]. These terms include charged particles both within the system and in the surroundings. The mutual energies of the particles within the system, in the absence of an external field, are macroscopically regarded as part of the internal energy [103]. Furthermore, by electric neutrality of the system $\rho_f = \rho_b = 0$, the volume average of all electric fields generated by such charges must be zero.

More specifically [18], in the lack of an external field there still exists a nonzero microscopic field energy density. The question is, how is this energy accounted for in the macroscopic continuum description? This energy cannot be viewed as a mere shift in the zero level of internal energy because it is dependent on interparticle distances and is therefore, in general, density and temperature dependent. Thus, this energy effectively contributes to changes in internal energy and must be included in the thermodynamic internal energy of the system [18].

The mutual energies of the external particles are not part of the Hamiltonian, as these interactions are no more of interest than that of the heat bath in thermal systems. The Hamiltonian includes all Coulomb terms associated with the energy stored within the system and the interaction energy of the external field with the dielectric body. The electric work done on the dielectric medium is part of the mutual energies of the external/internal interactions. Where do these external/internal energies belong? The surprising answer is that the microscopic Hamiltonian places the entire energy within the system [103]. Therefore the macroscopic treatment must also do so in order to be compatible [103].

The term $\mathcal{W}_0$ in (6.40) represents the mutual energy density of the external charges, in the presence of an infinite two-phase dielectric composite of laminates parallel to the applied field (see Section 1.2.1). The analysis done in Section 1.2.1 demonstrates that the electric field is uniform throughout the system, and is therefore independent of any charges within the dielectric body. Moreover, the interaction does not induce charge densities, and therefore performs no work on the medium. Consequently, this term is not included in the Hamiltonian nor the electric work term.

The term $\mathcal{W}_f$ is analogous to the pairwise self interaction term of the Ising model. It represents the mutual self-energy density of the dielectric body in its state of polarization, induced by the external charges. This term is included in the system Hamiltonian.

The term $\mathcal{W}_{int}$ represents the external charges interacting with, and polarizing a homogeneous dielectric medium with electric susceptibility $-(G(t;\alpha_\omega)-p/t)$. The above discussion of the term $\mathcal{W}_0$ illustrates why the contribution p/t is removed from this external/internal interaction term. One may be inclined to partition this interaction energy between the system and the surroundings, but the previous discussion has already indicated that it must be regarded as part of the system energy to be compatible with the microscopic description [103]. This interaction polarizes the dielectric body, changing the characteristic energy levels of the system, and determines the electric work done on the dielectric medium by the external charges. Consequently, it is this term that is to be placed in the first law for two-phase dielectric media (6.18), and is included in the Hamiltonian. In summary, the system Hamiltonian $\mathcal{H}_\omega$ is determined by $\mathcal{W}_{int}+\mathcal{W}_f=\frac{1}{2}\mathcal{W}_{int}$ and the electric work term δW is determined by $\mathcal{W}_{int}$.

In the interaction term $\mathcal{W}_{int}$, the quantity p/t represents a mere shift in the zero energy level of the system. This additive term is independent of the geometric configuration ω and can therefore be absorbed into the normalization of the probability measure. We will omit this contribution in the system Hamiltonian and the first law of thermodynamics. Results relative to the original basal level are recovered via the mapping $G(t;\alpha_\omega)\mapsto G(t;\alpha_\omega)-p/t$.

In order to make natural connections to physics, we define the following quantities

$$\chi^*_\omega(t)=-G(t;\alpha_\omega),\quad P^*_\omega(E_0,t)=\epsilon_1\chi^*_\omega(t)E_0,\quad \mathrm{p}^*_\omega(E_0,t)=\frac{P^*_\omega(E_0,t)}{N}\,. \tag{6.42}$$

For two-phase dielectric media, χ^*_ω plays the role of the effective electric susceptibility, P^*_ω plays the role of the effective polarization, p^*_ω plays the role of the effective dipole moment, and N^{-1} has units of volume. For matrix particle systems, N represents the number density of ϵ_2 inclusions, e.g., the number density of spheres in an ER fluid. We will use the language of matrix particle systems from now on, however our analysis holds for general two-phase stationary dielectric media in lattice and continuum settings [65].

We identify the permittivity of the matrix and particles by ϵ_1 and ϵ_2, respectively. We assume that the permittivity of the matrix is held fixed, and that it is *independent* of the temperature T. Metal particles are modeled by letting $\epsilon_2 \longrightarrow \infty$ $(t \longrightarrow 0)$ [74]. When the inclusions are identical particles, such as spheres in an ER fluid, the number density may be expressed by $N = p/V_p$ where p is the volume fraction of the particles and V_p is the volume of a single particle. Therefore N introduces a length scale into the problem.

A classical problem of electrostatics shows [66, 74] that the work done by external charges to initially polarize a homogeneous dielectric medium of permittivity ϵ^*_ω is $-\mathrm{p}^*_\omega E_0$. Including this energy contribution in the Hamiltonian $\mathcal{H}_\omega$, the above analysis shows that

$$\mathcal{H}_\omega = -\frac{\epsilon_1 E_0^2}{2N}\chi^*_\omega(t) = -\frac{1}{2}\mathrm{p}^*_\omega(E_0, t)E_0. \tag{6.43}$$

Furthermore, the work term to be inserted in the first law (6.18) of thermodynamics for two-phase dielectric media is given by

$$W = -\frac{\epsilon_1 E_0^2}{N}\langle\chi^*_\omega(t)\rangle_{\mathcal{H}} = -\mathrm{p}^*(T, E_0, t)E_0, \tag{6.44}$$

where $\langle\cdot\rangle_{\mathcal{H}}$ means GB canonical ensemble averaging. By the parameter separation property of the Hamiltonian and the first law (6.18), $d\mathcal{F} = -SdT - \mathrm{p}^* dE_0 - \Psi dt$, we have

$$\mathrm{p}^*(T, E_0, t) = -\frac{\partial \mathcal{F}}{\partial E_0}, \quad \mathcal{F} = -\beta^{-1}\ln Z, \quad Z = \sum_\omega \exp(-\beta\mathcal{H}_\omega). \tag{6.45}$$

Regardless of the physical arguments leading to (6.43) and (6.44), these equations define an especially convenient statistical mechanics model for two-phase dielectric media. In Theorem 11, we will show that the predictions of the model are consistent with experimental observations of ER fluids, and that the model is both physically transparent and mathematically tractable.

6.2.2 Maxwell's Relations and Stability

In this section we explore the consequences of Maxwell's relations in (6.18)-(6.20), and the physical consistency and stability of our model. Maxwell's relations provide representations for the state functions which are explicitly given in terms of the energy components of Theorem 4, and inherit the beautiful analytic properties of the system Hamiltonian. The main result of this chapter is given by Theorem 11.

Theorem 11 *The system Hamiltonian $\mathcal{H}_\omega$ and electric work term given in equations* (6.43) *and* (6.44)*, respectively, define a statistical mechanics model of the purely electric aspects of two-phase dielectric media with the following properties.*

(1) For each $\omega \in \Omega$, $\mathcal{H}_\omega$ is an is an analytic function of E_0, and t, for all $t < 0$, and is uniformly bounded for all $t < -\varepsilon < 0$. Moreover, in the infinite volume limit, $V \longrightarrow \infty$, the model captures all complicated geometric interactions exactly.

(2) The Electric components of the state functions are determined by the GB statistics of Herglotz functions involving the family of spectral measures $\{\alpha_\omega\}_{\omega\in\Omega}$, and are explicitly given in terms of the energy decomposition of Theorem 4. For example, the electric component S_e of the entropy S is determined by the GB variance of $\chi^*_\omega(t) = |G(t;\alpha_\omega)| = |\langle \chi_2 \vec{E} \cdot \vec{E}_0 \rangle / t E_0^2|$.

(3) The positivity of the entropy, $S(T, E_0, t) > 0$, is equivalent to

$$0 \leq \int_0^{E_0} (E_0')^3 \mathrm{Var}(\chi^*_\omega(t))_\mathcal{H} dE_0' < (2N^2 k/\epsilon_1^2)\, T^2\, S_0(T,t),$$

where $S_0(T,t) = S(T,0,t)$. Let $\mathcal{T} = \{(T,t) \in \mathbb{R}^+ \otimes \mathbb{R}^- : S_0(T,t) < \infty\}$, where $\mathbb{R}^+ = (0,\infty)$ and $\mathbb{R}^- = (-\infty, 0)$. Then, for all $(T,t) \in \mathcal{T}$, the GB variance of $\mathcal{H}_\omega$ vanishes as $E_0 \longrightarrow \infty$. Specifically, for all $(T,t) \in \mathcal{T}$, there exists a constant $\delta > 0$ such that

$$\mathrm{Var}(\chi^*_\omega(t))_\mathcal{H} = o(E_0^{-(4+\delta)}), \quad \mathrm{Var}(\mathcal{H}_\omega)_\mathcal{H} = o(E_0^{-\delta}), \quad E_0 \gg 1.$$

Furthermore, let $\mathcal{T}_0 = \{t \in \mathbb{R}^- : \lim_{T \longrightarrow 0} S_0(T,t) < \infty\}$. Then, for all $E_0 > 0$ and $t \in \mathcal{T}_0$, the GB variance of $\mathcal{H}_\omega$ vanishes as $T \longrightarrow 0$. Specifically, for all $t \in \mathcal{T}_0$, $\mathrm{Var}(\chi^*_\omega(t))_\mathcal{H} = o(T^2)$, as $T \longrightarrow 0$.

(6) Consistent with experimental observations of ER fluids, the GB average of the effective permittivity $\langle \epsilon^*_\omega \rangle_\mathcal{H}$ increases with E_0 and levels off as $E_0 \longrightarrow \infty$,

$$\partial \langle \epsilon^*_\omega \rangle_\mathcal{H} / \partial E_0 > 0, \qquad \partial \langle \epsilon^*_\omega \rangle_\mathcal{H} / \partial E_0 = o(E_0^{-(3+\delta)}), \quad E_0 \gg 1.$$

(5) The entropy decreases with E_0, $\partial S/\partial E_0 < 0$, a necessary condition for solidification in ER fluids.

(6) The model is locally stable in E_0 and T.

We will prove Theorem 11 via a sequence of lemmas.

Proof: Property (1) of Theorem 11 has already been established in Section 2.1. More specifically, $\mathcal{H}_\omega = (\epsilon_1 E_0^2/2N)\chi_\omega^*(t)$ is clearly analytic in E_0, and it was shown in [65] that $\chi_\omega^*(t) = -G(t;\alpha_\omega)$ an analytic function of t for all $t < 0$, and is uniformly bounded for all $t < -\varepsilon < 0$ [89]. Moreover, an ergodic theorem was given in [65] which proves that, in the infinite volume limit, the model captures all complicated geometric interactions *exactly*.

In order to make the proof of property (2) more transparent, we first prove some preliminary lemmas.

Lemma 14 *Let g_ω and h_ω be uniformly bounded random variables for $\omega \in \Omega$, and define $\Delta g_\omega = g_\omega - \langle g_\omega\rangle_{\mathcal{H}}$. Then the GB covariance $Cov(h_\omega, g_\omega)_{\mathcal{H}} = \langle h_\omega \Delta g_\omega\rangle_{\mathcal{H}}$ and $\langle h_\omega \Delta g_\omega \Delta h_\omega\rangle_{\mathcal{H}}$ satisfy*

$$(a)\ \ \langle h_\omega \Delta g_\omega\rangle_{\mathcal{H}} = \langle g_\omega \Delta h_\omega\rangle_{\mathcal{H}}, \quad (b)\ \ \langle h_\omega \Delta g_\omega \Delta h_\omega\rangle_{\mathcal{H}} = \langle h_\omega^2 \Delta g_\omega\rangle_{\mathcal{H}} - \langle h_\omega \Delta g_\omega\rangle_{\mathcal{H}}\langle h_\omega\rangle_{\mathcal{H}}$$

Proof: Properties (a) and (b) follow from

$$\langle h_\omega \Delta g_\omega\rangle_{\mathcal{H}} = \langle h_\omega(g_\omega - \langle g_\omega\rangle_{\mathcal{H}})\rangle_{\mathcal{H}} = \langle h_\omega g_\omega\rangle_{\mathcal{H}} - \langle h_\omega\rangle_{\mathcal{H}}\langle g_\omega\rangle_{\mathcal{H}} = \langle g_\omega h_\omega\rangle_{\mathcal{H}} - \langle g_\omega\rangle_{\mathcal{H}}\langle h_\omega\rangle_{\mathcal{H}},$$
$$\langle h_\omega \Delta g_\omega \Delta h_\omega\rangle_{\mathcal{H}} = \langle h_\omega \Delta g_\omega (h_\omega - \langle h_\omega\rangle_{\mathcal{H}})\rangle_{\mathcal{H}} = \langle h_\omega^2 \Delta g_\omega\rangle_{\mathcal{H}} - \langle h_\omega \Delta g_\omega\rangle_{\mathcal{H}}\langle h_\omega\rangle_{\mathcal{H}}, \ \square$$

Lemma 15 *Let $f_\omega = Z^{-1}\exp(\beta_e \chi_\omega^*(t))$, where $\beta_e = \beta\epsilon_1 E_0^2/2N$, and $\Delta\chi_\omega^* = \chi_\omega^* - \langle\chi_\omega^*\rangle_{\mathcal{H}}$. Then for $\xi = E_0, T$*

$$(a)\ \ \frac{\partial f_\omega}{\partial \xi} = \frac{\partial \beta_e}{\partial \xi} f_\omega \Delta\chi_\omega^*, \qquad (b)\ \ \frac{\partial f_\omega}{\partial t} = \beta_e f_\omega \Delta \frac{\partial \chi_\omega^*}{\partial t}$$

Proof:

$$\begin{aligned}
\text{(a)}\quad \frac{\partial f_\omega}{\partial \xi} &= \frac{\partial}{\partial \xi} Z^{-1}\exp(\beta_e\chi_\omega^*(t)) = \frac{\partial \beta_e}{\partial \xi}\chi_\omega^* f_\omega - Z^{-2}\exp(\beta_e\chi_\omega^*(t))\frac{\partial Z}{\partial \xi} \\
&= \frac{\partial \beta_e}{\partial \xi}\chi_\omega^* f_\omega - f_\omega Z^{-1}\sum_{\omega\in\Omega}\frac{\partial \beta_e}{\partial \xi}\chi_\omega^*\exp(\beta_e\chi_\omega^*(t)) = \frac{\partial \beta_e}{\partial \xi} f_\omega \Delta\chi_\omega^* \quad \square. \\
\text{(b)}\quad \frac{\partial f_\omega}{\partial t} &= \frac{\partial}{\partial t} Z^{-1}\exp(\beta_e\chi_\omega^*) = \beta_e\frac{\partial \chi_\omega^*}{\partial t} f_\omega - Z^{-2}\exp(\beta_e\chi_\omega^*(t))\frac{\partial Z}{\partial t} \\
&= \beta_e\frac{\partial \chi_\omega^*}{\partial t} f_\omega - f_\omega Z^{-1}\sum_{\omega\in\Omega}\beta_e\frac{\partial \chi_\omega^*}{\partial t}\exp(\beta_e\chi_\omega^*) = \beta_e f_\omega \Delta\frac{\partial \chi_\omega^*}{\partial t} \quad \square.
\end{aligned}$$

Lemma 16 shows that the mixed derivative commutators of the free energy $\mathcal{F}$ are zero, which establishes Maxwell's relations in (6.18)-(6.20).

Lemma 16 *Let* $\mathcal{F} = -\beta^{-1}\ln Z$*,* $Z = \sum_\omega \exp(\beta_e \chi^*_\omega(t))$*, and* $\beta_e = \beta\epsilon_1 E_0^2/2N$*. Define the mixed derivative commutator by* $[C_{x,y}] = [(\partial^2/\partial x\partial y) - (\partial^2/\partial y\partial x)]$*. Then*

$$(a)\;\; [C_{E_0,t}]\mathcal{F} = 0, \qquad (b)\;\; [C_{T,t}]\mathcal{F} = 0. \qquad (c)\;\; [C_{E_0,T}]\mathcal{F} = 0,$$

Proof: Properties (a)-(b) follow from Lemma 15, yielding

$$\begin{aligned}
\frac{\partial^2\mathcal{F}}{\partial t\partial\xi} &= \frac{\partial}{\partial t}\left[-\frac{\partial\beta^{-1}}{\partial\xi}\ln Z - \beta^{-1}\frac{\partial\beta_e}{\partial\xi}\langle\chi^*_\omega\rangle_\mathcal{H}\right] \\
&= \left[-\frac{\partial\beta^{-1}}{\partial\xi}\beta_e\left\langle\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_\mathcal{H} - \beta^{-1}\frac{\partial\beta_e}{\partial\xi}\left(\left\langle\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_\mathcal{H} + \beta_e\left\langle\chi^*_\omega\Delta\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_\mathcal{H}\right)\right] \\
&= -\frac{\partial}{\partial\xi}(\beta^{-1}\beta_e)\left\langle\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_\mathcal{H} - \beta^{-1}\beta_e\frac{\partial\beta_e}{\partial\xi}\left\langle\chi^*_\omega\Delta\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_\mathcal{H} \\
\frac{\partial^2\mathcal{F}}{\partial\xi\partial t} &= \frac{\partial}{\partial\xi}\left[-\beta^{-1}\beta_e\left\langle\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_\mathcal{H}\right] = -\frac{\partial}{\partial\xi}(\beta^{-1}\beta_e)\left\langle\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_\mathcal{H} - \beta^{-1}\beta_e\frac{\partial\beta_e}{\partial\xi}\left\langle\chi^*_\omega\Delta\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_\mathcal{H},
\end{aligned}$$

where $\xi = E_0, T$ □. Property (c) similarly follows by

$$\begin{aligned}
\frac{\partial^2\mathcal{F}}{\partial E_0\partial T} &= \frac{\partial}{\partial E_0}\left[-\frac{\partial\beta^{-1}}{\partial T}\ln Z - \beta^{-1}\frac{\partial\beta_e}{\partial T}\langle\chi^*_\omega\rangle_\mathcal{H}\right] \\
&= -\frac{\partial\beta^{-1}}{\partial T}\frac{\partial\beta_e}{\partial E_0}\langle\chi^*_\omega\rangle_\mathcal{H} - \beta^{-1}\frac{\partial^2\beta_e}{\partial E_0\partial T}\langle\chi^*_\omega\rangle_\mathcal{H} - \beta^{-1}\frac{\partial\beta_e}{\partial T}\frac{\partial\beta_e}{\partial E_0}\langle\chi^*_\omega\Delta\chi^*_\omega\rangle_\mathcal{H} \\
&= -\frac{\partial}{\partial T}\left(\beta^{-1}\frac{\partial\beta_e}{\partial E_0}\right)\langle\chi^*_\omega\rangle_\mathcal{H} - \beta^{-1}\frac{\partial\beta_e}{\partial T}\frac{\partial\beta_e}{\partial E_0}\langle\chi^*_\omega\Delta\chi^*_\omega\rangle_\mathcal{H} \\
\frac{\partial^2\mathcal{F}}{\partial T\partial E_0} &= \frac{\partial}{\partial T}\left[-\beta^{-1}\frac{\partial\beta_e}{\partial E_0}\langle\chi^*_\omega\rangle_\mathcal{H}\right] = -\frac{\partial}{\partial T}\left(\beta^{-1}\frac{\partial\beta_e}{\partial E_0}\right)\langle\chi^*_\omega\rangle_\mathcal{H} - \beta^{-1}\frac{\partial\beta_e}{\partial E_0}\frac{\partial\beta_e}{\partial T}\langle\chi^*_\omega\Delta\chi^*_\omega\rangle_\mathcal{H}\ \square.
\end{aligned}$$

Lemma 17 will be used in the analysis of Maxwell's relations.

Lemma 17 *Let* $\langle\chi^*_\omega\rangle_\mathcal{H} = Z^{-1}\sum_\omega \chi^*_\omega\exp(\beta_e\chi^*_\omega)$*,* $\beta_e = \beta\epsilon_1 E_0^2/2N$*,* $\Delta\chi^*_\omega = \chi^*_\omega - \langle\chi^*_\omega\rangle_\mathcal{H}$*, and* $[C_{x,y}] = [(\partial^2/\partial x\partial y) - (\partial^2/\partial y\partial x)]$*. Then, for* $\xi = E_0, T$

$$(a)\;\; [C_{E_0,T}]\langle\chi^*_\omega\rangle_\mathcal{H} = 0, \qquad (b)\;\; [C_{\xi,t}]\langle\chi^*_\omega\rangle_\mathcal{H} = \beta_e\frac{\partial\beta_e}{\partial\xi}\langle\chi^*_\omega\rangle_\mathcal{H}\,Cov\left(\chi^*_\omega, \frac{\partial\chi^*_\omega}{\partial t}\right)_\mathcal{H}$$

Proof: Property (a) follows from Lemma 14 and Lemma 15:

$$\begin{aligned}
\frac{\partial^2\langle\chi^*_\omega\rangle_\mathcal{H}}{\partial T\partial E_0} &= \frac{\partial}{\partial T}\left[\frac{\partial\beta_e}{\partial E_0}\langle\chi^*_\omega\Delta\chi^*_\omega\rangle_\mathcal{H}\right] \\
&= \frac{\partial^2\beta_e}{\partial T\partial E_0}\langle\chi^*_\omega\Delta\chi^*_\omega\rangle_\mathcal{H} + \frac{\partial\beta_e}{\partial E_0}\frac{\partial\beta_e}{\partial T}\langle\chi^*_\omega\Delta\chi^*_\omega\Delta\chi^*_\omega\rangle_\mathcal{H} \\
\frac{\partial^2\langle\chi^*_\omega\rangle_\mathcal{H}}{\partial E_0\partial T} &= \frac{\partial}{\partial E_0}\left[\frac{\partial\beta_e}{\partial T}\langle\chi^*_\omega\Delta\chi^*_\omega\rangle_\mathcal{H}\right] \\
&= \frac{\partial^2\beta_e}{\partial E_0\partial T}\langle\chi^*_\omega\Delta\chi^*_\omega\rangle_\mathcal{H} + \frac{\partial\beta_e}{\partial T}\frac{\partial\beta_e}{\partial E_0}\langle\chi^*_\omega\Delta\chi^*_\omega\Delta\chi^*_\omega\rangle_\mathcal{H}.
\end{aligned}$$

Therefore, as $[C_{E_0,T}]\beta_e = 0$ we have $[C_{E_0,T}]\langle\chi^*_\omega\rangle_{\mathcal{H}} = 0$ □. Properties (b)-(c) similarly follow from Lemma 14 and Lemma 15. For $\xi = E_0, T$,

$$\begin{aligned}
\frac{\partial^2\langle\chi^*_\omega\rangle_{\mathcal{H}}}{\partial\xi\partial t} &= \frac{\partial}{\partial\xi}\left[\left\langle\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_{\mathcal{H}} + \beta_e\left\langle\chi^*_\omega\Delta\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_{\mathcal{H}}\right] \\
&= \frac{\partial\beta_e}{\partial\xi}\left[2\left\langle\frac{\partial\chi^*_\omega}{\partial t}\Delta\chi^*_\omega\right\rangle_{\mathcal{H}} + \beta_e\left\langle\chi^*_\omega\Delta\frac{\partial\chi^*_\omega}{\partial t}\Delta\chi^*_\omega\right\rangle_{\mathcal{H}}\right] \\
\frac{\partial^2\langle\chi^*_\omega\rangle_{\mathcal{H}}}{\partial t\partial\xi} &= \frac{\partial}{\partial t}\left[\frac{\partial\beta_e}{\partial\xi}\langle\chi^*_\omega\Delta\chi^*_\omega\rangle_{\mathcal{H}}\right] = \frac{\partial\beta_e}{\partial\xi}\frac{\partial}{\partial t}\left(\left\langle(\chi^*_\omega)^2\right\rangle_{\mathcal{H}} - \langle\chi^*_\omega\rangle^2_{\mathcal{H}}\right) \\
&= \frac{\partial\beta_e}{\partial\xi}\left[\left\langle 2\chi^*_\omega\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_{\mathcal{H}} + \beta_e\left\langle(\chi^*_\omega)^2\,\Delta\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_{\mathcal{H}}\right] \\
&\quad - \frac{\partial\beta_e}{\partial\xi}\left[2\langle\chi^*_\omega\rangle_{\mathcal{H}}\left(\left\langle\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_{\mathcal{H}} + \beta_e\left\langle\chi^*_\omega\Delta\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_{\mathcal{H}}\right)\right] \\
&= \frac{\partial\beta_e}{\partial\xi}\left[2\left\langle\chi^*_\omega\Delta\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_{\mathcal{H}} + \beta_e\left\langle\chi^*_\omega\Delta\frac{\partial\chi^*_\omega}{\partial t}\Delta\chi^*_\omega\right\rangle_{\mathcal{H}}\right] \\
&\quad - \beta_e\frac{\partial\beta_e}{\partial\xi}\langle\chi^*_\omega\rangle_{\mathcal{H}}\left\langle\chi^*_\omega\Delta\frac{\partial\chi^*_\omega}{\partial t}\right\rangle_{\mathcal{H}}
\end{aligned}$$

The result now follows from Lemma 14 □. We are now ready to establish property (2) of Theorem 11.

We now show that first law, $d\mathcal{F} = -SdT - \mathrm{p}^*dE_0 - \Psi dt$, and Maxwell's relations in (6.18) explicitly determine the state functions. Indeed, by equation (6.44), and Lemma 16, we have

$$\frac{\partial S}{\partial E_0} = \frac{\partial\mathrm{p}^*}{\partial T} = \frac{\epsilon_1 E_0}{N}\,\frac{\partial\langle\chi^*_\omega\rangle_{\mathcal{H}}}{\partial T}, \qquad \frac{\partial\Psi}{\partial E_0} = \frac{\partial\mathrm{p}^*}{\partial t} = \frac{\epsilon_1 E_0}{N}\,\frac{\partial\langle\chi^*_\omega\rangle_{\mathcal{H}}}{\partial t}. \tag{6.46}$$

Therefore, if we denote by S_0 and Ψ_0 the entropy and contrast potential when $E_0 = 0$, respectively, then integrating (6.46) with respect to E_0 yields

$$\begin{aligned}
S(T,E_0,t) &= S_0(T,t) + S_e(T,E_0,t), \qquad S_e(T,E_0,t) = \int_0^{E_0}\frac{\epsilon_1 E_0'}{N}\,\frac{\partial\langle\chi^*_\omega\rangle_{\mathcal{H}}}{\partial T}\,dE_0', \qquad (6.47)\\
\Psi(T,E_0,t) &= \Psi_0(T,t) + \Psi_e(T,E_0,t), \qquad \Psi_e(T,E_0,t) = \int_0^{E_0}\frac{\epsilon_1 E_0'}{N}\frac{\partial\langle\chi^*_\omega\rangle_{\mathcal{H}}}{\partial t}dE_0',
\end{aligned}$$

where S_e and Ψ_e are the *electric components* of the entropy and contrast potential, respectively. The third of Maxwell's relations in (6.18), $\partial S/\partial t = \partial\Psi/\partial T$, then yields the following lemma

Lemma 18

$$Cov\left(\chi^*_\omega, \frac{\partial\chi^*_\omega}{\partial t}\right)_{\mathcal{H}} \equiv 0, \tag{6.48}$$

independent of the values of the state variables T*,* E_0*, and* t*, and the details of the underlying random medium* (Ω, P).

Proof: *Equations* (6.18) *and* (6.47), *and Lemma 17 imply that*

$$\frac{\partial S_0}{\partial t} - \frac{\partial \Psi_0}{\partial T} = \int_0^{E_0} \frac{\epsilon_1 E_0'}{N} \, [C_{T,t}] \langle \chi_\omega^* \rangle_{\mathcal{H}} \, dE_0'$$
$$= \int_0^{E_0} \frac{\epsilon_1 E_0'}{N} \, \beta_e \, \frac{\partial \beta_e}{\partial T} \, \langle \chi_\omega^* \rangle_{\mathcal{H}} \; Cov\left(\chi_\omega^*, \frac{\partial \chi_\omega^*}{\partial t}\right)_{\mathcal{H}} dE_0' \, .$$

The E_0 *independence of* S_0 *and* Ψ_0 *implies that there exists a constant c, say, such that*

$$\frac{\partial S_0}{\partial t} - \frac{\partial \Psi_0}{\partial T} = c = \int_0^{E_0} \frac{\epsilon_1 E_0'}{N} \, \beta_e \frac{\partial \beta_e}{\partial T} \langle \chi_\omega^* \rangle_{\mathcal{H}} \; Cov\left(\chi_\omega^*, \frac{\partial \chi_\omega^*}{\partial t}\right)_{\mathcal{H}} dE_0'$$

Since $(\epsilon_1 E_0' \beta_e / N)(\partial \beta_e / \partial T) \not\equiv 0$ *and* $\langle \chi_\omega^* \rangle_{\mathcal{H}} > 0$ $(\chi_\omega^* > 0$ *for all* $\omega \in \Omega)$, *the condition* $\partial c / \partial E_0 = 0$ *yields equation* (6.48), *independent of the values of the state variables* T, E_0, *and* t, *and the details of the underlying medium.*

Lemma 18 has two immediate corollaries.

Corollary 2

(a) $[C_{E_0,T}] \langle \chi_\omega^* \rangle_{\mathcal{H}} = 0$, *(b)* $[C_{E_0,t}] \langle \chi_\omega^* \rangle_{\mathcal{H}} = 0$, *(c)* $[C_{T,t}] \langle \chi_\omega^* \rangle_{\mathcal{H}} = 0$.

Corollary 2 follows directly from Lemma 17 and Lemma 18.

Corollary 3

$$\frac{\partial}{\partial t} \langle \chi_\omega^* \rangle_{\mathcal{H}} = \left\langle \frac{\partial \chi_\omega^*}{\partial t} \right\rangle_{\mathcal{H}}$$

Corollary 3 follows directly from Lemma 15 and Lemma 18.

Lemma 15 and Corollary 3 shows that the electric components S_e and Ψ_e of the entropy and contrast potential in (6.47) are given by

$$S_e(T, E_0, t) = \int_0^{E_0} \frac{\epsilon_1 E_0'}{N} \, \frac{\partial \beta_e}{\partial T} \, \mathrm{Var}(\chi_\omega^*)_{\mathcal{H}} \, dE_0', \quad \Psi_e(T, E_0, t) = \int_0^{E_0} \frac{\epsilon_1 E_0'}{N} \left\langle \frac{\partial \chi_\omega^*}{\partial t} \right\rangle dE_0'. \tag{6.49}$$

We note that, as $\partial \beta / \partial T = -\beta / T < 0$, the electric component of the entropy S_e is negative $S_e < 0$. Recalling that $\chi_\omega^*(t) = -G(t; \alpha_\omega)$, the decomposition of system energy given in Theorem 4 shows that

$$\chi_\omega^* = -\int_0^1 \frac{d\alpha_\omega(\lambda)}{t - \lambda} = -\frac{\langle \chi_2 \vec{E} \cdot \vec{E}_0 \rangle}{t E_0^2} > 0, \qquad \frac{\partial \chi_\omega^*}{\partial t} = \int_0^1 \frac{d\alpha_\omega(\lambda)}{(t - \lambda)^2} = \frac{\langle \chi_2 E^2 \rangle}{t^2 E_0^2} > 0. \tag{6.50}$$

Therefore the electric components S_e and Ψ_e of these state functions are determined by GB statistics of Herglotz functions involving the family of spectral measures $\{\alpha_\omega\}_{\omega \in \Omega}$, and are explicitly given in terms of the energy decomposition of Theorem 4. This establishes property (2) of Theorem 11.

The remainder of Maxwell's relations in (6.19) and (6.20) can be expressed in terms of the logarithmic functional $\Lambda(T, E_0, t) = \ln \langle \chi^*_\omega \rangle_{\mathcal{H}}$. A perturbation analysis shows that these relations constitute a detailed balance of energy in the system.

Lemma 19

$$\begin{aligned} &(a) \quad -\frac{\partial E_0}{\partial t} = \frac{\partial \Psi}{\partial p^*} = E_0 \frac{\partial \Lambda}{\partial t}\left(1 + E_0 \frac{\partial \Lambda}{\partial E_0}\right)^{-1}, \\ &(b) \quad -\frac{\partial E_0}{\partial T} = \frac{\partial S}{\partial p^*} = E_0 \frac{\partial \Lambda}{\partial T}\left(1 + E_0 \frac{\partial \Lambda}{\partial E_0}\right)^{-1}, \\ &(c) \quad -\frac{\partial t}{\partial T} = \frac{\partial S}{\partial \Psi} = \frac{\partial \Lambda}{\partial T}\left(\frac{\partial \Lambda}{\partial E_0}\right)^{-1}. \end{aligned} \tag{6.51}$$

Proof: Property (a) follows from the properties of partial differentiation [103], and equations (6.19), (6.20), (6.44), and (6.46), yielding

$$\begin{aligned} -\frac{\partial E_0}{\partial t} &= \frac{\partial \Psi}{\partial \mathrm{p}^*} = \frac{\partial \Psi}{\partial E_0}\left(\frac{\partial \mathrm{p}^*}{\partial E_0}\right)^{-1} = \frac{\partial \mathrm{p}^*}{\partial t}\left(\frac{\partial \mathrm{p}^*}{\partial E_0}\right)^{-1} \\ &= \frac{\epsilon_1 E_0}{N}\frac{\partial \langle \chi^*_\omega \rangle_{\mathcal{H}}}{\partial t}\left(\frac{\epsilon_1}{N}\langle \chi^*_\omega \rangle_{\mathcal{H}} + \frac{\epsilon_1 E_0}{N}\frac{\partial \langle \chi^*_\omega \rangle_{\mathcal{H}}}{\partial E_0}\right)^{-1} = E_0 \frac{\partial \Lambda}{\partial t}\left(1 + E_0 \frac{\partial \Lambda}{\partial E_0}\right)^{-1}, \end{aligned}$$

which are, in turn given by the energy components in (6.50). Similarly,

$$\begin{aligned} -\frac{\partial E_0}{\partial T} &= \frac{\partial S}{\partial \mathrm{p}^*} = \frac{\partial S}{\partial E_0}\left(\frac{\partial \mathrm{p}^*}{\partial E_0}\right)^{-1} = \frac{\partial \mathrm{p}^*}{\partial T}\left(\frac{\partial \mathrm{p}^*}{\partial E_0}\right)^{-1} \\ &= \frac{\epsilon_1 E_0}{N}\frac{\partial \langle \chi^*_\omega \rangle_{\mathcal{H}}}{\partial T}\left(\frac{\epsilon_1}{N}\langle \chi^*_\omega \rangle_{\mathcal{H}} + \frac{\epsilon_1 E_0}{N}\frac{\partial \mathrm{p}^*}{\partial E_0}\right)^{-1} = E_0 \frac{\partial \Lambda}{\partial T}\left(1 + E_0 \frac{\partial \Lambda}{\partial E_0}\right)^{-1} \end{aligned}$$

Finally,

$$\begin{aligned} -\frac{\partial t}{\partial T} &= \frac{\partial S}{\partial \Psi} = \frac{\partial S}{\partial E_0}\left(\frac{\partial \Psi}{\partial E_0}\right)^{-1} = \frac{\partial \mathrm{p}^*}{\partial T}\left(\frac{\partial \mathrm{p}^*}{\partial t}\right)^{-1} \\ &= \frac{\epsilon_1 E_0}{N}\frac{\partial \langle \chi^*_\omega \rangle_{\mathcal{H}}}{\partial T}\left(\frac{\epsilon_1 E_0}{N}\frac{\partial \langle \chi^*_\omega \rangle_{\mathcal{H}}}{\partial t}\right)^{-1} = \frac{\partial \Lambda}{\partial T}\left(\frac{\partial \Lambda}{\partial E_0}\right)^{-1}. \end{aligned}$$

It is worth mentioning that through Padé approximants $G_n(t;\alpha_\omega) = P_n^{[1]}(t,\omega)/P_n(t,\omega)$ of $G(t;\alpha_\omega)$ (see section Section 4.1), we have, with exponential convergence,

$$\ln\left(G_n(t,\omega)\right) = \ln\left(\frac{P_n^{[1]}(t,\omega)}{P_n(t,\omega)}\right) = \int_0^1 \ln(t-\lambda)[d\rho_n^{[1]} - d\rho_n](\lambda,\omega), \tag{6.52}$$

where $\rho_n^{[1]}$ and ρ_n are the root distributions of the polynomials $P_n^{[1]}(t,\omega)$ and $P_n(t,\omega)$, respectively. This suggests that the logarithmic functional Λ may also have a representation similar to (6.52), with respect to a signed measure.

Now we establish property (3) of Theorem 11. Recall that $\beta_e = \beta\epsilon_1 E_0^2/2N$, where $\beta = 1/kT$. Equations (6.47) and (6.49), and $\partial\beta/\partial T = -\beta/T$ imply that the positivity of the entropy, $S(T, E_0, t) > 0$, is equivalent to

$$0 \leq \int_0^{E_0} (E_0')^3 \mathrm{Var}(\chi_\omega^*(t))_{\mathcal{H}} dE_0' < (2N^2k/\epsilon_1^2)\, T^2\, S_0(T,t), \tag{6.53}$$

Define the sets $\mathbb{R}^+ = (0,\infty)$, $\mathbb{R}^- = (-\infty, 0)$, $\mathcal{T} = \{(T,t) \in \mathbb{R}^+ \otimes \mathbb{R}^- : S_0(T,t) < \infty\}$, and $\mathcal{T}_0 = \{t \in \mathbb{R}^- : \lim_{T\longrightarrow 0} S_0(T,t) < \infty\}$. Then, analogous to Theorem 10, (6.53) implies that, for all $(T,t) \in \mathcal{T}$, the GB variance of $\mathcal{H}_\omega$ vanishes as $E_0 \longrightarrow \infty$. More specifically, for all $(T,t) \in \mathcal{T}$, there exists a constant $\delta > 0$ such that

$$\mathrm{Var}(\chi_\omega^*(t))_{\mathcal{H}} = o(E_0^{-(4+\delta)}), \quad \mathrm{Var}(\mathcal{H}_\omega)_{\mathcal{H}} = o(E_0^{-\delta}), \quad E_0 \gg 1. \tag{6.54}$$

Furthermore, for all $t \in \mathcal{T}_0$, $E_0 > 0$, equation (6.53) implies that the GB variance of $\mathcal{H}_\omega$ vanishes as $T \longrightarrow 0$. More specifically, $\mathrm{Var}(\chi_\omega^*(t))_{\mathcal{H}} = o(T^2)$, as $T \longrightarrow 0$, for all $t \in \mathcal{T}_0$.

Property (4) of Theorem 11 also follows from equation (6.54). It implies that the GB average of the effective permittivity $\langle\epsilon_\omega^*\rangle_{\mathcal{H}} = \epsilon_1(1 + \langle\chi_\omega^*\rangle_{\mathcal{H}})$ increases with E_0 and levels off as $E_0 \longrightarrow \infty$:

$$\begin{aligned}
&\frac{\partial\langle\epsilon_\omega^*\rangle_{\mathcal{H}}}{\partial E_0} = \epsilon_1 \frac{\partial\langle\chi_\omega^*\rangle_{\mathcal{H}}}{\partial E_0} = \epsilon_1 \frac{\partial\beta_e}{\partial E_0} \mathrm{Var}(\chi_\omega^*(t))_{\mathcal{H}} = \frac{\beta\epsilon_1^2 E_0}{N} \mathrm{Var}(\chi_\omega^*(t))_{\mathcal{H}} > 0, \\
&\frac{\partial\langle\epsilon_\omega^*\rangle_{\mathcal{H}}}{\partial E_0} = o(E_0^{-(3+\delta)}), \qquad E_0 \gg 1.
\end{aligned}$$

We have, en route, already established property (5) of Theorem 11, which follows from $\partial\beta/\partial T = -\beta/T$, equation (6.46), and Lemma 15, which imply

$$\frac{\partial S}{\partial E_0} = \frac{\epsilon_1 E_0}{N} \frac{\partial\langle\chi_\omega^*\rangle_{\mathcal{H}}}{\partial T} = \frac{\epsilon_1 E_0}{N} \frac{\partial\beta_e}{\partial T} \mathrm{Var}(\chi_\omega^*(t))_{\mathcal{H}} = \frac{\epsilon_1 E_0}{N} \left(-\frac{\beta\epsilon_1 E_0^2}{2NT}\right) \mathrm{Var}(\chi_\omega^*(t))_{\mathcal{H}} < 0. \tag{6.55}$$

We now establish property (6) of Theorem 11. For thermodynamic stability, it is necessary that the free energy $\mathcal{F}$ be a concave function of its intensive parameters T, E_0, and t, and a convex function of its extensive ones [103]. These conditions are restricted by the differential based analysis given in this section, which apply only to the *local* shape of the potential $\mathcal{F}$. If the full potential surface resembles a hilly terrain, it is possible that there could be neighboring deeper valleys or higher peaks that offer improved stability. The following calculation confirms property (6) of Theorem 11. By equations (6.44) and (6.49), and Lemma 15 we have

$$-\frac{\partial^2 \mathcal{F}}{\partial E_0^2} = \frac{\partial \mathrm{p}^*}{\partial E_0} = \frac{\partial}{\partial E_0}\frac{\epsilon_1 E_0}{N}\langle \chi^*_\omega \rangle_{\mathcal{H}} = \frac{\epsilon_1}{N}\left(\langle \chi^*_\omega \rangle_{\mathcal{H}} + E_0 \frac{\partial \beta_e}{\partial E_0}\mathrm{Var}(\chi^*_\omega)_{\mathcal{H}}\right) > 0 \tag{6.56}$$

$$-\frac{\partial^2 \mathcal{F}}{\partial T^2} = \frac{\partial S}{\partial T} = \frac{\partial S_0}{\partial T} + \int_0^{E_0} \frac{\epsilon_1 E_0'}{N}\frac{\partial}{\partial T}\left[\frac{\partial \beta_e}{\partial T}\mathrm{Var}(\chi^*_\omega)_{\mathcal{H}}\right] dE_0'$$

$$= \frac{\partial S_0}{\partial T} + \int_0^{E_0} \frac{\epsilon_1 E_0'}{N}\left[\frac{\partial^2 \beta_e}{\partial T^2}\mathrm{Var}(\chi^*_\omega)_{\mathcal{H}} + \left(\frac{\partial \beta_e}{\partial T}\right)^2 \langle \chi^*_\omega \Delta\chi^*_\omega \Delta\chi^*_\omega \rangle_{\mathcal{H}}\right] dE_0' > 0,$$

where $\langle \chi^*_\omega \Delta\chi^*_\omega \Delta\chi^*_\omega \rangle_{\mathcal{H}} = \langle \chi^*_\omega (\Delta\chi^*_\omega)^2 \rangle_{\mathcal{H}} > 0$ and we have assumed the local stability of the thermodynamic system when $E_0 = 0$: $\partial S_0/\partial T > 0$. The positivity of all other terms has already been discussed above. This concludes Theorem 11 $\square$.

In this chapter, we reviewed the information theory approach to statistical mechanics, yielding the canonical ensemble. In Theorem 10, we characterized the behavior of this ensemble by the deviation of its statistics, away from that of the underlying reference measure, toward that of a probability measure which is highly localized around minimum energy states. The Hamiltonian of our model is given by $\mathcal{H}_\omega = -(\epsilon_1 E_0^2/2N)\chi^*_\omega$. Therefore, the GB probability measure is precisely the reference measure $P(d\omega)$ when $E_0 = 0$. Theorem 10 suggests that, as E_0 increases and/or T decreases, the statistics of the ensemble should tend toward that of a probability measure which is highly localized around *maximum* χ^*_ω states. This is indeed the behavior of our model, which is indicated by parts (3) and (6) of Theorem 11. They illustrate that, as E_0 increases, the GB probability measure becomes increasingly localized around states $\omega \in \Omega$ that maximize $\langle \epsilon^*_\omega \rangle_{\mathcal{H}} = \epsilon_1(1 + \langle \chi^*_\omega \rangle_{\mathcal{H}})$, i.e., maximize $\langle \chi^*_\omega \rangle_{\mathcal{H}}$, and as $E_0 \longrightarrow \infty$, $\mathrm{Var}(\chi^*_\omega)_{\mathcal{H}} \longrightarrow 0$. This behavior is consistent with experimental observations of ER fluids [126, 128]. In future work we will further explore the predictions of this model, including the existence of a phase transition.

Figure 6.1 displays an ordered sequence of configurations of the square 2-d bond network, with increasing ϵ^*_{xx}, the x component of the effective permittivity. As ϵ^*_{xx} increases, the system orders itself into increasingly connected structures, elongated in the x direction. This is analogous to an ER fluid with average electric field $\vec{E}_0 = E_0\vec{e}_x$, where $\vec{e}_x$ is a unit vector pointing in the x direction. Similarly, Figure 6.2 displays an ordered sequence of configurations of the square 2-d bond network, with increasing ϵ^*_{yy}. This is analogous to an ER fluid with average electric field $\vec{E}_0 = E_0\vec{e}_y$, and as ϵ^*_{yy} increases, the system orders itself into increasingly connected structures, elongated in the y direction. Finally Figure 6.3 displays an ordered sequence of configurations of the square 2-d bond network,

Figure 6.1. An ordered sequence of configurations of the square 2-d bond network, with increasing ϵ^*_{xx}, the x component of the effective permittivity. As ϵ^*_{xx} increases, from left to right and top to bottom, the system orders itself into increasingly connected structures, elongated in the x direction.

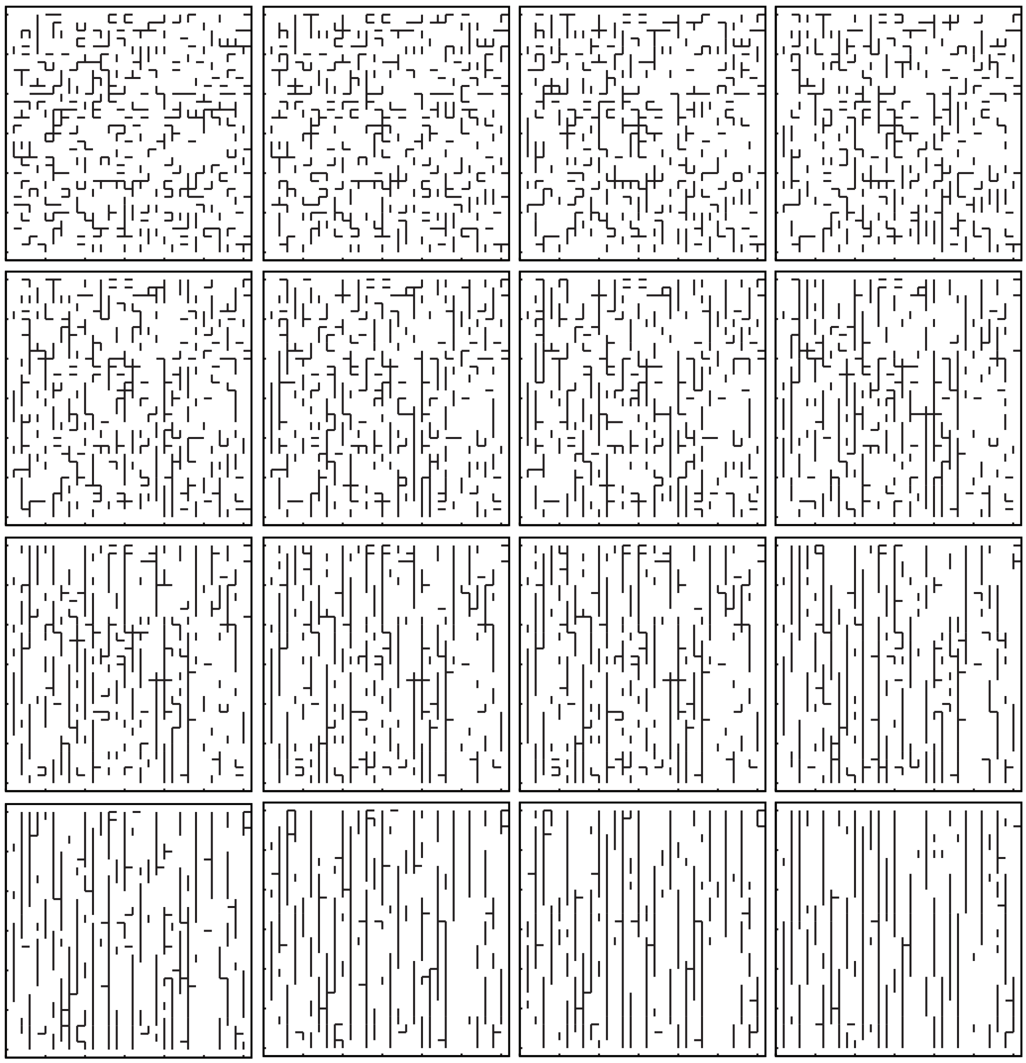

Figure 6.2. An ordered sequence of configurations of the square 2-d bond network, with increasing ϵ^*_{yy}, the y component of the effective permittivity. As ϵ^*_{yy} increases, from left to right and top to bottom, the system orders itself into increasingly connected structures, elongated in the y direction.

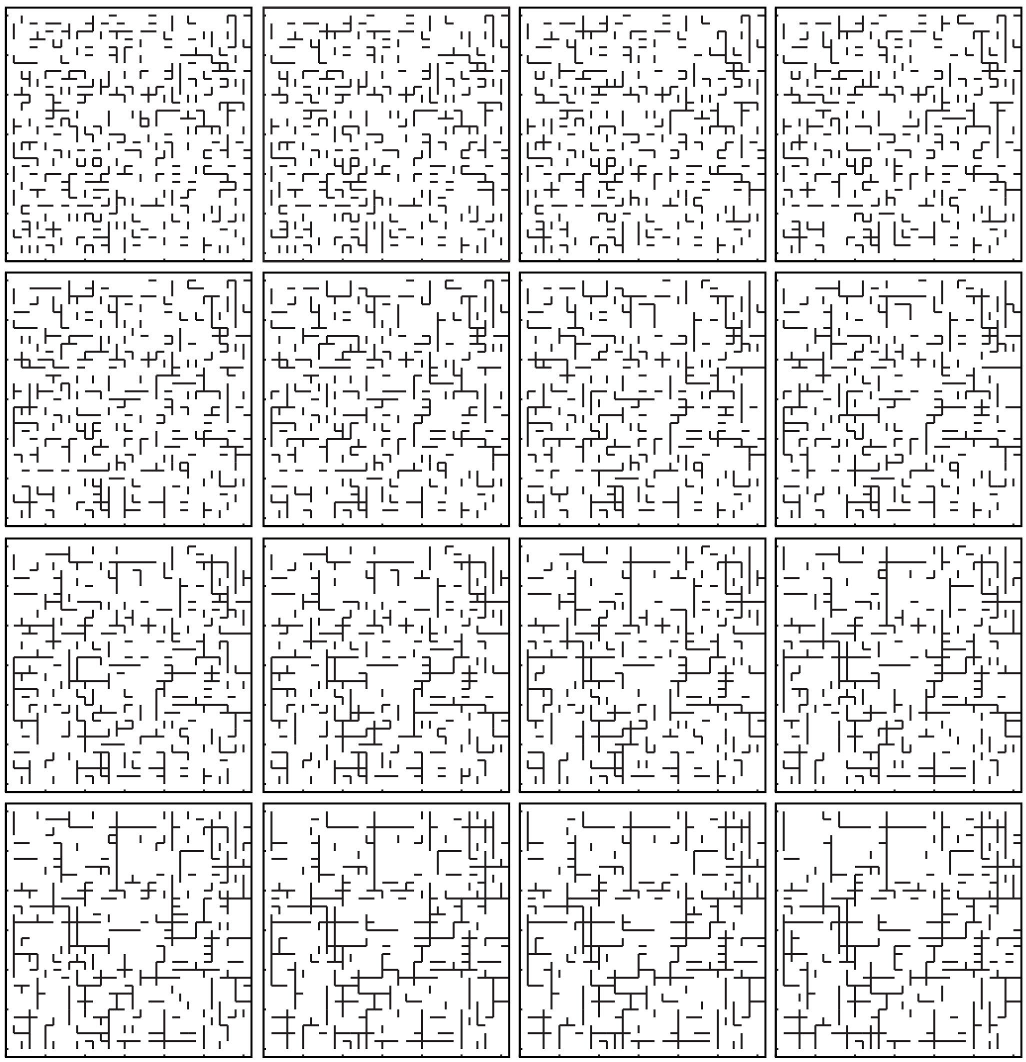

Figure 6.3. An ordered sequence of configurations of the square 2-d bond network, with increasing $\epsilon^*_{xx} + \epsilon^*_{yy}$, the trace of the effective permittivity tensor. As $\epsilon^*_{xx} + \epsilon^*_{yy}$ increases, from left to right and top to bottom, the system orders itself into increasingly connected structures.

with increasing $\epsilon^*_{xx} + \epsilon^*_{yy}$, the trace of the effective permittivity tensor $\boldsymbol{\epsilon}^*$. As $\epsilon^*_{xx} + \epsilon^*_{yy}$ increases, the system orders itself into increasingly connected fractal-like structures. This is analogous to an ER fluid with metallic inclusions.

These figures were generated by directly calculating the components of the tensor valued spectral measure $\boldsymbol{\alpha}$ in (3.39), from the random matrix $M_2 = \chi_2 \Gamma \chi_2$, by the method outlined in Section 3.3. The calculation of $\boldsymbol{\alpha}$ yields the effective permittivity tensor $\boldsymbol{\epsilon}^*$ for the network. The first frame of these figures is a randomly generated configurations of the bonds. The evolution of the system is determined as follows. Let us focus on ϵ^*_{xx} as the procedure involving ϵ^*_{yy} and $\epsilon^*_{xx} + \epsilon^*_{yy}$ is identical. Let ω_0 denote the initial configuration of the bonds. The x component $\alpha_{xx}(\omega_0)$ of the spectral measure is calculated from the matrix $M_2(\omega_0)$ associated with this initial configuration. This yields $\epsilon^*_{xx}(\omega_0)$ via $G(t; \alpha_{xx}(\omega_0)) = \sum_{j=1}^n m_j/(t - \lambda_j)$, where the m_j are defined directly below equation (3.39) and $t = 1/(1 - \epsilon_2/\epsilon_1) = -1.8$. Another random configuration ω_1 is then generated and $\epsilon^*_{xx}(\omega_1)$ is computed in the same manner. If $\epsilon^*_{xx}(\omega_1) > \epsilon^*_{xx}(\omega_0)$ then the move is accepted. Otherwise it is rejected and the process repeats. These simulations indicate, by spectral means, that equilibrium configurations of ER fluids are *local* states of maximum $\epsilon^* = \boldsymbol{\epsilon}^* \vec{e}_0 \cdot \vec{e}_0$, where $\vec{E}_0 = E_0 \vec{e}_0$, and further validate our model.

We conclude this chapter by noting a possible extension of this statistical mechanics model to our critical theory discussed in Chapter 3. There, we studied critical behavior exhibited by percolation models of two-phase conductive media, as a function of the volume fraction p of the σ_2 phase. In such models, as p increases, the system becomes increasingly connected, and there is a critical transition in the transport properties of the effective conductivity σ^* as p surpasses the percolation threshold p_c.

In ER fluids, it is E_0 that controls the connectedness of the system. Experiments have shown [70, 120, 128] that, as E_0 increases, the system becomes increasingly connected, and there is a critical transition in the transport properties of the effective permittivity ϵ^* as E_0 surpasses a critical value E_c. From equation (2.18) of Section 2.2, we have $\hat{g}(h; \hat{\phi}_\omega) = (t-1)G(t; \alpha_\omega) = (1-t)\chi^*_\omega(t)$, in the variables $h = \epsilon_1/\epsilon_2$ and $t = h/(h-1)$, and metal particles are modeled by letting $h \longrightarrow 0$ ($\epsilon_2 \longrightarrow \infty,\ 0 < \epsilon_1 < \infty$) [74]. Therefore, assuming a Fubini theorem, equation (6.45) implies that

$$
\begin{aligned}
-\frac{\partial \mathcal{F}}{\partial E_0} &= \mathrm{p}^*(T, E_0, t) = \frac{\epsilon_1 E_0}{N} \langle \chi_\omega^* \rangle_{\mathcal{H}} = \frac{\epsilon_1 E_0}{N(1-t)} \left\langle \int_0^\infty \frac{d\hat{\phi}_\omega(y)}{1+hy} \right\rangle_{\mathcal{H}} \\
&= \frac{\epsilon_1 E_0}{N(1-t)} \int_0^\infty \frac{\langle d\hat{\phi}_\omega(y) \rangle_{\mathcal{H}}}{1+hy} \, .
\end{aligned}
$$

Recall that the fundamental assumption of our critical theory is the following. For $p < p_c$, there is a "gap" θ_α about $\lambda = 0$ which shrinks as p increases and the system becomes increasingly connected, and $\theta_\alpha \longrightarrow 0$ as $p \longrightarrow p_c^-$. In the case of ER fluids, assume that, for $E_0 < E_c$, there is a gap θ_α about $\lambda = 0$ which shrinks as E_0 increases and the system becomes increasingly connected, and $\theta_\alpha \longrightarrow 0$ as $E_0 \longrightarrow E_c^-$. With this parallel in the language of critical these transitions, under the substitution $\hat{g}(h; \hat{\phi}) \mapsto \hat{g}(h; \langle \hat{\phi}_\omega \rangle_{\mathcal{H}})$, the ideas underlying Theorem 5 extend to our statistical mechanics model of ER fluids, with metallic particles. However, recall that the measure $\hat{\phi}$ is independent of the contrast parameter h, and the measure $\langle \hat{\phi}_\omega \rangle_{\mathcal{H}}$ is dependent on h, E_0, and T, in a highly nonlinear way. Therefore the proof of such an extension requires some care. This is a key part of our future work.

REFERENCES

[1] A. Y. Abul-Magd. Non-extensive random matrix theory — a bridge connecting chaotic and regular dynamics. *Phys. Lett. A*, 333(1-2):16–22, 2004.

[2] A. Y. Abul-Magd. Non-extensive random-matrix theory based on Kaniadakis entropy. *Phys. Lett. A*, 361(6):450–454, 2007.

[3] G. A. Baker. Some rigorous inequalities satisfied by the ferromagnetic Ising model in a magnetic field. *Phys. Rev. Lett.*, 20:990–992, 1968.

[4] G. A. Baker. *Quantitative Theory of Critical Phenomena*. Academic Press, New York, 1990.

[5] G. A. Baker and D. S. Gaunt. Low-temperature critical exponents from high-temperature series: The Ising model. *Phys. Rev. B*, 1(3):1184–1210, 1970.

[6] R. Balian. Random matrices and information theory. *Nuovo Cimento B*, 57(183):471–475, 1968.

[7] J. N. Bandyopadhyay and S. Jalan. Universality in complex networks: random matrix analysis. *Phys. Rev. E*, 76:026109–1–4, 2007.

[8] C. W. J. Beenakker and B. Rejaei. Random-matrix theory of parametric correlations in the spectra of disordered metals and chaotic billiards. *Phys. A*, 203(1):61–90, 1994.

[9] V. L. Berdichevsky. *Thermodynamics of Chaos and Order*. Addison Wesley Longman Inc., Edinburgh, UK, 1997.

[10] D. J. Bergman. The dielectric constant of a composite material – A problem in classical physics. *Phys. Rep. C*, 43(9):377–407, 1978.

[11] D. J. Bergman. Exactly solvable microscopic geometries and rigorous bounds for the complex dielectric constant of a two-component composite material. *Phys. Rev. Lett.*, 44:1285–1287, 1980.

[12] D. J. Bergman. Rigorous bounds for the complex dielectric constant of a two–component composite. *Ann. Phys.*, 138:78–114, 1982.

[13] D. J. Bergman and D. Stroud. Physical properties of macroscopically inhomogeneous media. *Solid State Phys.*, 46:147–269, 1992.

[14] L. Berlyand and K. Golden. Exact result for the effective conductivity of a continuum percolation model. *Phys. Rev. B*, 50(4):2114–2117, July 1994.

[15] M. V. Berry and J. P. Keating. The Riemann zeros and eigenvalue asymptotics. *SIAM Rev.*, 41:236–266, 1999.

[16] C. Blecken and K. A. Muttalib. Brownian motion model of a q-deformed random matrix ensemble. *J. Phys. A: Math. Gen.*, 31:2123–2132, 1998.

[17] R. Blumenfeld, Y. Meir, A. Aharony, and A. B. Harris. Resistance fluctuations in randomly diluted networks. *Phys. Rev. B*, 35:3524–3535, 1987.

[18] S. Bobbio. *Electrodynamics of Materials*. Academic Press, San Diego, CA, 2000.

[19] C. Bonifasi-Lista and E. Cherkaev. Electrical impedance spectroscopy as a potential tool for recovering bone porosity. *Phys. Med. Biol.*, 54(10):3063–3082, 2009.

[20] T. Bourbie and B. Zinszner. Hydraulic and acoustic properties as a function of porosity in Fontainebleau sandstone. *J. Geophys. Res.*, 90(B13):11,524–11,532, 1985.

[21] S. R. Broadbent and J. M. Hammersley. Percolation processes I. Crystals and mazes. *Proc. Cambridge Philos. Soc.*, 53:629–641, 1957.

[22] T. A. Brody, J. Flores, J. B. French, P. A. Mello, A. Pandey, and S. S. M. Wong. Random-matrix physics: Spectrum and strength fluctuations. *Rev. Modern Phys.*, 53(3):385–480, 1981.

[23] O. Bruno. The effective conductivity of strongly heterogeneous composites. *Proc. R. Soc. London A*, 433:353–381, 1991.

[24] O. Bruno and K. Golden. Interchangeability and bounds on the effective conductivity of the square lattice. *J. Stat. Phys.*, 61:365, 1990.

[25] H. B. Callen. *Thermodynamics and an Introduction to Thermostatistics*. Wiley, New York, 1985.

[26] C. M. Canali. Model for a random-matrix description of the energy-level statistics of disordered systems at the Anderson transition. *Phys. Rev. B*, 53(7):3713–3730, 1996.

[27] D. Chandler. *Introduction to Modern Statistical Mechanics*. Oxford University Press, Oxford, 1987.

[28] Y. Chen, M. E. H. Ismail, and K. A. Muttalib. A solvable random matrix model for disordered conductors. *J. Phys., Condens. Matter*, 71(4):L417–L423, 1992.

[29] Y. Chen, M. E. H. Ismail, and K. A. Muttalib. Metallic and insulating behavior in an exactly solvable random matrix model. *J. Phys., Condens. Matter*, 71(5):177–190, 1993.

[30] Y. Chen and C. A. Schuh. Percolation of diffusional creep: a new universality class. *Phys. Rev. Lett.*, 98:035701–1–4, 2007.

[31] E. Cherkaev. Inverse homogenization for evaluation of effective properties of a mixture. *Inverse Problems*, 17(4):1203–1218, 2001.

[32] E. Cherkaev and C. Bonifasi-Lista. Characterization of structure and properties of bone by spectral measure method. *J. Biomech.*, 44(2):345 – 351, 2011.

[33] E. Cherkaev and K. M. Golden. Inverse bounds for microstructural parameters of composite media derived from complex permittivity measurements. *Waves in Random Media*, 8(4):437–450, 1998.

[34] E. Cherkaev and M.-J. Ou. De-homogenization: Reconstruction of moments of the spectral measure of composite. *Inverse Problems*, 24:065008 (19pp), 2008.

[35] T. S. Chihara. *An Introduction to Orthogonal Polynomials*. Gordon and Breach, New York, 1978.

[36] K. Christensen and N. R. Moloney. *Complexity and Criticality*. Imperial College Press, Covent Garden, London, 2005.

[37] J. P. Clerc, G. Giraud, J. M. Laugier, and J. M. Luck. The electrical conductivity of binary disordered systems, percolation clusters, fractals, and related models. *Adv. Phys.*, 39(3):191–309, 1990.

[38] C. A. Davis, D. R. McKenzie, and R. C. McPhedran. Optical properties and microstructure of thin silver films. *Optics Comm.*, 85(1):70–82, 1991.

[39] A. R. Day and M. F. Thorpe. The spectral function of random resistor networks. *J. Phys.: Cond. Matt.*, 8:4389–4409, 1996.

[40] A. R. Day and M. F. Thorpe. The spectral function of composites: the inverse problem. *J. Phys.: Cond. Matt.*, 11:2551–2568, 1999.

[41] P. Deift. *Orthogonal Polynomials and Random Matrices: a Riemann–Hilbert Approach*. Courant Institute of Mathematical Sciences, New York, NY, 2000.

[42] P. Deift. *Random Matrix Theory: Invariant Ensemble and Universality*. Courant Institute of Mathematical Sciences, New York, NY, 2009.

[43] P. Deift and D. Gioev. Universality at the edge of the spectum for unitary, orthogonal, and symplectic ensembles of random matrices. *Comm. Pure Appl. Math.*, 60(6):867–910, 2007.

[44] P. Deift, L. C. Li, and C. Tomei. Toda flows with infinitely many variables. *J. Funct. Anal.*, 64(3):358–402, 1985.

[45] E. Deuring, R. Blumenfeld, D. J. Bergman, A. Aharony, and M. Murat. Current distributions in a two-dimensional random-resistor network. *J. Stat. Phys.*, 3267:113–121, 1992.

[46] F. J. Dyson. A Brownian-motion model for the eigenvalues of a random matrix. *J. Math. Phys.*, 3(6):1191–1198, 1962.

[47] F. J. Dyson. Statistical theory of the energy levels of complex systems. I. *J. Math. Phys.*, 3:140–156, 1962.

[48] F. J. Dyson. Statistical theory of the energy levels of complex systems. II. *J. Math. Phys.*, 3:157–165, 1962.

[49] F. J. Dyson. Statistical theory of the energy levels of complex systems. III. *J. Math. Phys.*, 3:166–175, 1962.

[50] F. J. Dyson. A class of matrix ensembles. *J. Math. Phys.*, 13(1):90–97, 1972.

[51] F. J. Dyson and M. L. Mehta. Statistical theory of the energy levels of complex systems IV. *J. Math. Phys.*, 4(5):701–712, 1963.

[52] A. L. Efros and B. I. Shklovskii. Critical behavior of conductivity and dielectric constant near the metal–non-metal transition threshold. *Phys. Stat. Sol. (b)*, 76(2):475–485, 1976.

[53] I. G. Enting. A lattice statistics model for the age distribution of air bubbles in polar ice. *Nature*, 315:654–655, 1985.

[54] G. B. Folland. *Introduction to Partial Differential Equations*. Princeton University Press, Princeton, NJ, 1995.

[55] G. B. Folland. *Real Analysis: Modern Techniques and Their Applications*. Wiley–Interscience, New York, NY, 1999.

[56] P. J. Forrester. *Log-gases and random matrices*. Princeton University Press, Princeton, New Jersey, 2010.

[57] W. Gautshci. *Orthogonal Polynomials Computation and Approximation*. Oxford Press, Oxford New York, 2004.

[58] K. M. Golden. Exponent inequalities for the bulk conductivity of a hierarchical model. *Commun. Math. Phys.*, 143(3):467–499, 1992.

[59] K. M. Golden. Statistical mechanics of conducting phase transitions. *J. Math. Phys.*, 36(10):5627–5642, 1995.

[60] K. M. Golden. Critical behavior of transport in lattice and continuum percolation models. *Phys. Rev. Lett.*, 78(20):3935–3938, 1997.

[61] K. M. Golden. Climate change and the mathematics of transport in sea ice. *Notices Amer. Math. Soc.*, 56(5):562–584 and issue cover, 2009.

[62] K. M. Golden, S. F. Ackley, and V. I. Lytle. The percolation phase transition in sea ice. *Science*, 282:2238–2241, 1998.

[63] K. M. Golden, H. Eicken, A. L. Heaton, J. Miner, D. Pringle, and J. Zhu. Thermal evolution of permeability and microstructure in sea ice. *Geophys. Res. Lett.*, 34:L16501 (6 pages and issue cover), 2007.

[64] K. M. Golden, N. B. Murphy, and E. Cherkaev. Spectral analysis and connectivity of porous microstructures in bone. *J. Biomech.*, 44(2):337–344, 2011.

[65] K. M. Golden and G. Papanicolaou. Bounds for effective parameters of heterogeneous media by analytic continuation. *Commun. Math. Phys.*, 90:473–491, 1983.

[66] D. J. Griffiths. *Introduction to Electrodynamics*. Prentice Hall, Upper Saddle River, NJ, 1999.

[67] T. Guhr, A. Müller-Groeling, and H. A. Weidenmüller. Random-matrix theories in quandom physics: Common concepts. *Phys. Rept.*, 299(4):189–425, 1998.

[68] A. Gully, L. G. E. Backstrom, H. Eicken, and K. M. Golden. Complex bounds and microstructural recovery from measurements of sea ice permittivity. *Physica. B, Condensed matter*, 394(2):357–362, 2007.

[69] B. I. Halperin, S. Feng, and P. N. Sen. Differences between lattice and continuum percolation transport exponents. *Phys. Rev. Lett.*, 54(22):2391–2394, 1985.

[70] T. C. Halsey. Electrorheological fluids. *Science*, 258:761–766, 1992.

[71] P. Henrici. *Applied and Computational Complex Analysis. Volume 3.* John Wiley & Sons Inc., New York, 1974.

[72] D. C. Hong, H. E. Stanley, A. Coniglio, and A. Bunde. Random walk approach to the two-component random-conductor mixture: Perturbing away from the perfect random resistor network and random superconducting-network limits. *Phys. Rev. B*, 33:4564, 1986.

[73] M. E. H. Ismail. *Classical and Quantum Orthogonal Polynomials in One Variable.* Cambridge University Press, UK, 2005.

[74] J. D. Jackson. *Classical Electrodynamics.* John Wiley and Sons, Inc., New York, 1999.

[75] T. Jonckheere and J. M. Luck. Dielectric resonances of binary random networks. *J. Phys. A: Math. Gen.*, 31:3687–3717, 1998.

[76] V. E. Kravtsov and K. A. Muttalib. New class of random matrix ensembles with multifractal eigenvectors. *Phys. Rev. Lett.*, 79(10):1913–1916, 1997.

[77] R. Kühn. Spectra of sparse random matrices. *J. Phys. A: Math. Theor.*, 41(6):295002, 2008.

[78] R. P. Kusy. Influence of particle size ratio on the continuity of aggregates. *J. Appl. Phys.*, 48(12):5301–5303, 1977.

[79] R. P. Kusy and D. T. Turner. Electrical resistivity of a polymeric insulator containing segregated metallic particles. *Nature*, 229:58–59, 1971.

[80] A. V. Kyrylyuk and P. van der Schoot. Continuum percolation of carbon nanotubes in polymeric and colloidal media. *Proc. Nat. Acad. Sci. USA*, 105:8221–8226, 2008.

[81] T. D. Lee and C. N. Yang. Statistical theory of equations of state and phase transitions. II. Lattice gas and Ising model. *Phys. Rev.*, 87:410–419, 1952.

[82] F. Luo, J. Zhong, Y. Yang, R. H. Scheuermann, and J. Zhou. Application of random matrix theory to biological networks. *Phys. Lett. A*, 357:420–423, 2006.

[83] N. F. G. Martin and J. W. England. *Mathematical Theory of Entropy.* Addison Wesley, Reading, Mass., 1981.

[84] M. L. Mehta. *Random Matrices.* Elsevier, Amsterdam, third edition, 2004.

[85] F. Mezzadri and N. C. Snaith. *Recent Perspectives in Random Matrix Theory and Number Theory.* Cambridge University Press, Cambridge, UK, 2005.

[86] G. W. Milton. Bounds on the complex dielectric constant of a composite material. *Appl. Phys. Lett.*, 37:300–302, 1980.

[87] G. W. Milton. *Theory of Composites*. Cambridge University Press, Cambridge, 2002.

[88] N. B. Murphy. Random matrix characterization of phase transitions in two phase random media. In preparation., 2012.

[89] N. B. Murphy and K. M. Golden. The Ising model and critical behavior of transport in binary composite media. In press, J. Math. Phys., 39 pages., 2012.

[90] N. B. Murphy and K. M. Golden. Random matrix theory for composite materials. In preparation., 2012.

[91] N. B. Murphy, K. M. Golden, and P. Sheng. Statistical mechanics of homogenization for composite media. Preprint, 20 pages (in revision before submission)., 2012.

[92] N. B. Murphy, C. Hohenegger, C. S. Sampson, D. K. Perovich, H. Eicken, E. Cherkaev, B. Alali, and K. M. Golden. Spectral analysis of multiscale sea ice structures in the climate system. In preparation., 2012.

[93] K. A. Muttalib, Y. Chen, and M. E. H. Ismail. *q-Random Matrix Ensembles in "Symbolic computation, number theory, special functions, physics and combinatorics*, chapter 9, pages 1–23. Kluwer Academic, Amsterdam, Netherlands, 2001.

[94] K. A. Muttalib, Y. Chen, M. E. H. Ismail, and V. N. Nicopoulos. New family of unitary random matrices. *Phys. Rev. Lett.*, 71(4):471–475, 1993.

[95] C. Orum, E. Cherkaev, and K. M. Golden. Recovery of inclusion separations in strongly heterogeneous composites from effective property measurements. *Proc. Roy. Soc. London A*, 468(2139):784–809, 2012.

[96] B. N. Parlett. *The Symmetric Eigenvalue Problem*, volume 20 of *Classics in applied mathematics*. SIAM, Philidelphia, PA, Corrected reprint of the 1980 original.

[97] L. A. Pastur. From random matrices to quasi-periodic Jacobi matrices via orthogonal polynomials. *J. Approx. Theory*, 139(1):269–292, 2006.

[98] D. J. Pringle, J. E. Miner, H. Eicken, and K. M. Golden. Pore-space percolation in sea ice single crystals. *J. Geophys. Res. (Oceans)*, 114:C12017, 2009.

[99] K. Rajan and L. F. Abbott. Eigenvalue spectra of random matrices for neural networks. *Phys. Rev. Lett.*, 97(18):188104–1–188104–4, 2006.

[100] F. Rassoul-Agha and T. Seppäläinen. *A Course on Large Deviation Theory with an Introduction to Gibbs Measures*. Unpublished, 2008.

[101] M. C. Reed and B. Simon. *Functional Analysis*. Academic Press, San Diego CA, 1980.

[102] J. R. Reitz, F. J. Milford, and R. W. Christy. *Foundations of Electromagnetic Theory.* Addison–Wesley Publishing Company, San Francisco, CA, 1993.

[103] H. S. Robertson. *Statistical Thermophysics.* Prentice Hall, Englewood Cliffs, NJ, 1993.

[104] W. Rudin. *Real and Complex Analysis.* McGraw-Hill, Inc., New York, NY, 1987.

[105] D. Ruelle. *Statistical Mechanics: Rigorous Results.* W. A. Benjamin, New York, 1969.

[106] D. Ruelle. Extension of the Lee-Yang circle theorem. *Phys. Rev. Lett.*, 26(6):303–304, 1971.

[107] D. Ruelle. Characterization of Lee–Yang polynomials. *Ann. Math.*, 171:589–603, 2010.

[108] E. B. Saff and V. Totik. *Logarithmic Potentials with External Fields.* Springer, New York, 1997.

[109] N. Sasaki, H. Yamamura, and N. Matsushima. Is there a relation between bone strength and percolation? *J. Theor. Biol.*, 122(1):25–31, 1986.

[110] B. K. P. Scaife. *Principles of Dielectrics.* Clarendon Press, Oxford, 1989.

[111] J. P. Sethna. *Entropy, Order Parameters, and Complexity.* Oxford University Press, New York, 2006.

[112] P. Sheng and R. V. Kohn. Geometric effects in continuous-media percolation. *Phys. Rev. B*, 26:1331–1335, 1982.

[113] B. I. Shklovskii and A. L. Efros. *Electronic Properties of Doped Semiconductors.* Springer Verlag, New York, 1984.

[114] J. A. Shohat and J. D. Tamarkin. *The Problem of Moments.* American mathematical society, Providence, RI, 1963.

[115] B. Simon. Equilibrium measures and capacities in spectral theory. *Inverse Probl. Imag.*, 1(4):713–772, 2007.

[116] D. Stauffer and A. Aharony. *Introduction to Percolation Theory.* Taylor and Francis, London, second edition, 1992.

[117] A. D. Stone, P. A. Mello, K. A. Muttalib, and J. L. Pichard. *Random Matrix Theory and Maximum Entropy Models for Disordered Conductors*, chapter 9, pages 369–448. Elsevier Science Publishers, Amsterdam, Netherlands, 1991.

[118] M. H. Stone. *Linear Transformations in Hilbert Space.* American Mathematical Society, Providence, RI, 1964.

[119] G. Szegö. *Orthogonal Polynomials.* AMS, Providence, RI, 1939.

[120] R. Tao and G. D. Roy (eds.). *Electrorheological Fluids: Mechanisms, Properties, Technology, and Applications.* World Scientific, Singapore, 1994.

[121] C. J. Thompson. *Classical Equilibrium Statistical Mechanics.* Oxford University Press, Oxford, 1988.

[122] S. Torquato. *Random Heterogeneous Materials: Microstructure and Macroscopic Properties*. Springer-Verlag, New York, 2002.

[123] M. J. Uline, D. W. Siderius, and D. S. Corti. On the generalized equipartition theorem in molecular dynamics ensembles and the microcanonical thermodynamics of small systems. *J. Chem. Phys.*, 128:124301–1–124301–17, 2008.

[124] W. Van Assche. Orthogonal polynomials, associated polynomials and functions of the second kind. *J. Comp. Appl. Math.*, 37(1):237–249, 1991.

[125] W. Van Assche. Padé and Hermite–Padé approximation and orthogonality. *Surv. Approx. Theory*, 2(1):61–91, 2006.

[126] W. Wen, X. Huang, and P. Sheng. Electrorheological fluids: Structures and mechanisms. *Soft Matter*, 4(2):200–210, 2008.

[127] W. Wen and K. Lu. Frequency dependence of metal-particle/insulating oil electrorheological fluids. *Appl. Phys. Lett.*, 67:2147–2148, 1995.

[128] W. Wen and K. Lu. Pattern transitions induced by the surface properties of suspended microspheres in electrorheological fluid. *Phys. of Fluids*, 9:1826–1829, 1997.

[129] E. P. Wigner. On the statistical distribution of the widths and spacings of nuclear resonance levels. *Proc. Camb. Philos. Soc.*, 47(4):790–798, 1951.

[130] C. N. Yang and T. D. Lee. Statistical theory of equations of state and phase transitions. I. Theory of condensation. *Phys. Rev.*, 87:404–409, 1952.

[131] D. Zhang and E. Cherkaev. Reconstruction of spectral function from effective permittivity of a composite material using rational function approximations. *J. Comput. Phys.*, 228(15):5390 – 5409, 2009.