Conductivity and superconductivity in models of electron-phonon interaction

van Dijk, L

Rijks*universiteit* Groningen

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Luuk Adriaan Cornelis van Dijk

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Introduction

In der Beschränkung zeigt sich der Meister

It is the privilege of a physicist to be allowed to study a bewildering variety of phenomena in the world around us. With this privilege comes the obligation to aim at a deep understanding of these phenomena and hence the necessity to restrict oneself to a subset of them.

The subset of physics of the solid state is exemplary in many ways. Solid state systems have been compared to miniature universes[18] populated by elementary particles like electrons, phonons and magnons. Thus they are not only of interest in themselves but they also form a playground to study ideas originating from and applicable to other areas of physics, ranging from the physics of the Planck scale to cosmological phenomena. Moreover, the fruits born by solid state physics, with the transistor as the primary example, have changed everyday life on our planet in many ways in the course of this century.

Constructing theories is a two–step process: A model is constructed from ideas about reality (see fig.1), and then the model itself becomes a subject of study, to investigate the phenomena it actually predicts and to check the validity and relevance for the experiments it was supposed to describe. This thesis focuses on the interaction of electrons and phonons, ie. lattice vibrations in crystalline solids. On very general principles a theory may be constructed that should reproduce all aspects of this interplay. But no matter how general the principles, this theory would be so tremendously complicated as to be useless for any prediction.

The reason lies in the fact that a solid is a system composed of *very* many particles. To make the analysis feasible we would like to leave out as many unnecessary details as possible. For example: to calculate that a material crystallizes at a given pressure and temperature, starting from the model Hamiltonian of all the electrons and all the nuclei, is a tremendously difficult task which we may simply skip if we are interested in properties in the crystal itself and not in its formation. Also, we may assume that the electromagnetic field reacts very fast compared to the ions and electrons, giving rise to an effective force between electrons and ions that may be described in terms of two–body interactions, rather than the emission and absorption of photons by the massive particles. This way many degrees of freedom are removed from the Hamiltonian, and it is rendered in a compact form describing phonons, electrons moving over the lattice, and effective couplings between all these.

The Fröhlich and the Holstein model, introduced below, for the interaction between electrons and phonons are easy to write down, a property they share with the Hubbard model for effective electron–electron interactions, the Heisenberg model for magnetic interactions as well as various combinations and limits of these. These four models may be viewed as further approximations, each trying to cover a particular aspect of the wealth of phenomena present in the solid state[10, 30].

Polarons and Peierls states

In such an effective description we can investigate the electron–phonon interaction and the resulting electron–electron interaction that is mediated through the exchange of pho*n*ons rather than pho*t*ons. (We will ignore all magnetic aspects.) From a field–theoretical perspective one may say that whenever two matter fields interact their excitations may form bound states,

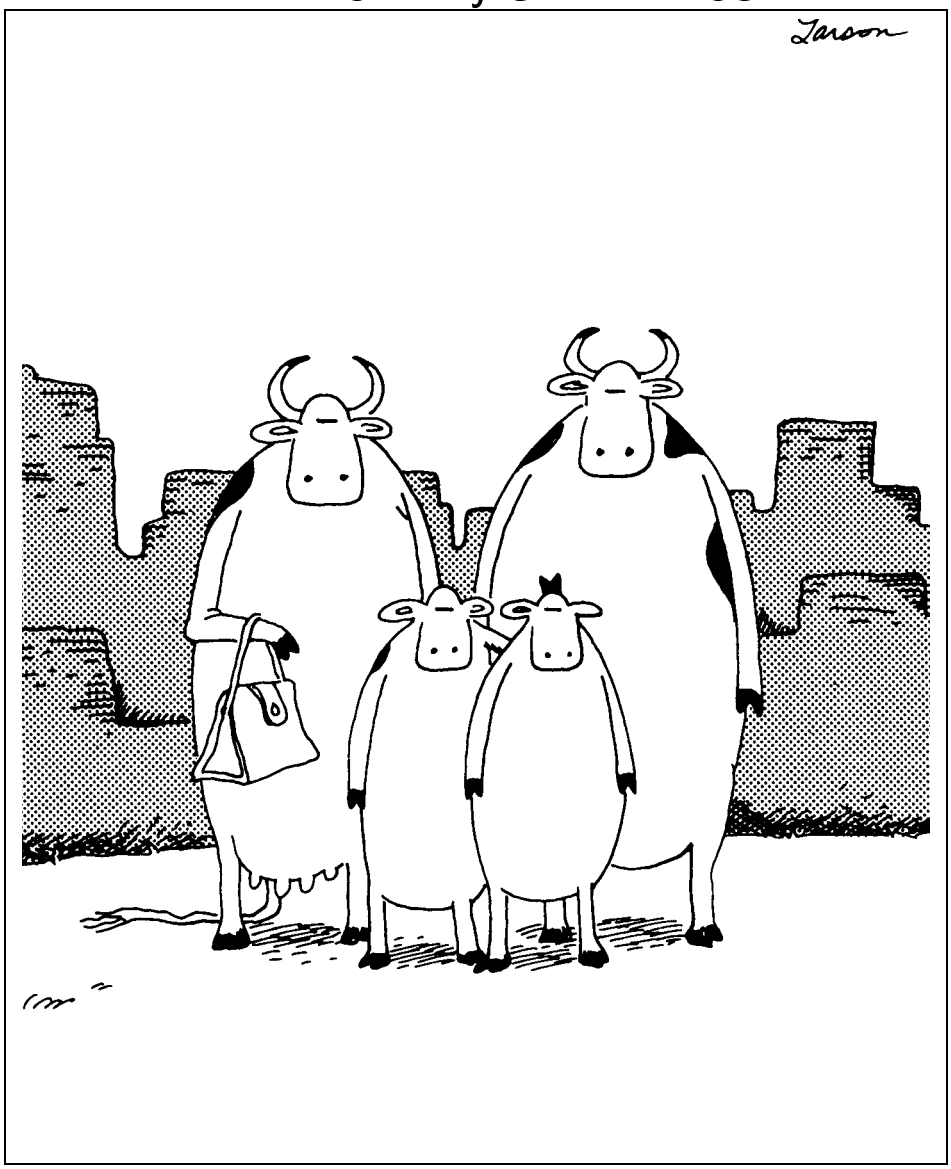

Figure 1: *Theoretical solid–state physicists state their models customarily in the form of a Hamiltonian, $\mathcal{H} = \ldots$. This is a peculiar professional deformation that may occasionally lead to surprises when confronted with other uses, scientific or everyday, of the word "model"[62].*

leading to composite particles which usually have their own name. In the case of electron and phonon fields the composite particle is called a polaron.

The following analogy for a polaron is due to Feynman: Imagine a ball on a mattress. Through its weight it creates a distortion in the mattress around itself. When it wants to move it has to take this distortion with it. This limits the transport of balls over the mattress. With the electron as the ball, the crystal lattice as the mattress, the weight of the ball as the interaction strength, polarisation as the distortion and conductivity as ball-transport, this is the theory of polarons in a nutshell. Two such balls may fall into each others dip and thus create a new bound state of two polarons, a *bi*polaron. When the binding of two electrons takes place in reciprocal space rather than in real space, we may expect the formation of Cooper pairs, which are responsible for superconductivity.

In contrast to these cases which can be understood in a picture of just one or two electrons that interact through the exchange of phonons, stands the example due to Peierls[18]. In a one–dimensional system where the electrons couple to the lattice, the distribution of electrons and the lattice may distort in a periodic way, with a period of two lattice sites. In this "Peierls state" the system is an insulator for electricity. This is a simple but non–trivial example of correlated behaviour of many particles.

From the above examples (that will reappear in this thesis) it should be clear that the effect of whatever goes on inside the material in terms of microscopic states and ordering will be noticed through its macroscopic transport properties, like electrical conductivity and superconductivity. These are among the key properties to be calculated for any proposed model.

Numerical Methods

The models that are obtained by reducing the description from the full many body system to just the degrees of freedom we consider to be relevant may be easy to write down, but despite this apparent simplicity, the study of their mathematical consequences like transport properties may be the work of a lifetime. To calculate the desired transport and ordering properties in general extra approximations have to be made, that are no longer *motivated* by physical principles but at best are *justified* by them. Many of these approximation schemes are perturbative, in the sense that they are an expansion in some small parameter, say (inverse) temperature, or some internal coupling[25]. These approximation methods have arisen naturally where analytic methods have run into infeasibility problems.

With the availability of fast and cheap computers, some of these borders of infeasibility have been pushed further back, whereas others remain as forbidding as ever. Rather than trying to proceed by analytic methods as far as possible before invoking numerical calculations, we take a step back and wonder if, given that we have to make approximations anyway, we may not make approximations that on one hand are physically as sound as traditional ones (or even sounder), but on the other hand are more efficient and accurate to implement numerically. These approximations need not be of a perturbative nature. Loosely speaking, we call a calculation "numerically exact" if we have an upper bound to the error in the outcome of the calculation, and this bound can be made to vanish in the limit of infinite amounts of CPU–cycles and computer memory available.

The method to study the class of models that is the subject of this thesis is numerically exact in a certain limit, and allows the calculation of a broad range of transport and ordering properties. In addition, a non–perturbative way is developed to check the validity of extrapolation of results from this limit to the non–limiting cases.

Having zoomed in from physics in general to the subject of this thesis: numerical calculation

of transport properties of models of electron–phonon interaction, the remainder of this introductory chapter will put the present work in the slightly broader and more technical perspective of the literature that has been devoted to the subject over the past six decades, and outline the contents of the remaining chapters.

Large and small polarons and superconductivity

Contemplating the effect of an electron on the polarisation of the surrounding lattice, Landau was the first to introduce the concept of polarons in the thirties of this century. Since then, the body of literature on this topic has become massive. For a thorough recent review see Devreese[23].

Fröhlich actually derived the Hamiltonian for this "large polaron" and treated the strong and weak coupling limits. Feynman in 1955 reformulated the model in the form of a path integral in which the phonon degrees of freedom could be integrated out[6]. Part of the motivation in those days was the hope of understanding the phenomena of superconductivity in terms of polarons. When BCS theory finally appeared, a few years after Fröhlich's and Feynman's work, this provided a microscopic answer to a riddle that had intrigued physicists for over five decades. The composite particle that turns out to be responsible for this phenomenon is not the polaron but the Cooper pair, consisting of two electrons of opposite spin and momentum, bound by exchanging phonons.

When the lattice distortion that accompanies an electron is confined to a region of the order of a lattice cell, one deals with a "small polaron". The Holstein model[20] was introduced by Holstein to study this limit, and will be the prime example model used in this thesis. The application Holstein had in mind was the Molecular Crystal. An alternate model, for the molecular crystal of trans–polyacetylene, is the SSH–model, introduced by Su, Schrieffer and Heeger[43], which is very similar, for the purpose of this thesis.

The work of Hirsch and Fradkin[48] and De Raedt and Lagendijk[24] in the beginning of the previous decade marks the advent of numerical methods to obtain new insights on previously inaccessible parts of the phase diagrams of these electron–phonon systems.

The discovery of the High–T_c superconductors in the mid eighties then raised many questions concerning the responsible mechanism and possible alternatives to the BCS theory. This renewed interest in models for polaronic mechanisms for superconductivity, especially the role of Fröhlich (bi)polarons in High–T_c superconductors[61, 69, 70, 71, 72], and of strongly correlated electrons[38] in general. Specific investigations included the competition between electron–electron Coulomb repulsion, as modeled in its simplest form by the Hubbard model, the phonon–mediated electron–electron attraction, as present in the Holstein model, and the magnetic exchange described by the 'J' of the t-J model. The combined Hubbard–Holstein, and Holstein–t-J models display rich phase diagrams which still leave a lot to be discovered[33, 34, 35, 36, 37].

We may view the topic of electron–phonon and related models as a space spanned by the characteristic energy scales of the subsystems involved: the electronic energy E_e , the phonon energy E_{ph}, the electron–phonon coupling energy $E_{e\text{-}ph}$ (Holstein and SSH models), the Coulomb interaction between electrons $E_{e\text{-}e}$ (Hubbard model), not to mention the magnetic interaction E_J (tJ-model etc.) Then there are the thermodynamic parameters of temperature and density, and aspects that cannot be parametrized like dimensionality and the lattice structure. Corners, edges, sides etcetera in this (at least seven–dimensional) space correspond to certain limits, eg. the adiabatic limit $E_{ph}/E_e \to 0$, the strong coupling limits $E_e/E_{e\text{-}ph} \to 0$ or $E_e/E_{e\text{-}e} \to 0$.

Apart from the methods used, analytic or numerical, and the properties calculated, publications can now be classified by the area they cover in this space. The rest of this thesis

will deal with a class of generalized Holstein–Fröhlich Hamiltonians from the perspective of computational physics. In terms of the parameters named above, we can classify this as taking place in the space of nonzero E_e, E_{ph} and $E_{e\text{-}ph}$, but for $E_{e\text{-}e} = E_J = 0$. The numerical method operates in the adiabatic limit, $E_{ph}/E_e \to 0$ and is independent of the details of the lattice structure and of the interaction, and works for any temperature and density. A big advantage over other methods found in the literature is the fact that dynamical properties (ie. transport properties) can be calculated in an efficient and straightforward way.

Sketch of contents

Chapter 1 will introduce the class of models, the notation and some tools used in this work.

The adiabatic limit will be introduced in chapter 2. It is this limit that is accessible to efficient numerical methods: thermodynamical averages can be evaluated in two steps: the electronic part can be done analytically, and the remaining phonon degrees of freedom can be integrated over by Monte Carlo methods. This approach must then be supplied with a tool to estimate the range where we may expect this limit to be a valid approximation for the non–limiting case. The primary tools for this are upper and lower bounds to the free energy. Also treated are some analytic results that allow for comparison between the full class, the limit and some special cases. Finally, the numerical recipe to simulate the model in the adiabatic limit on a computer is described and illustrated using the half–filled one–dimensional Holstein model at zero temperature.

Chapter 3 then treats a specific example of a transport property, the electric conductivity, that may be calculated from simulations. The results are compared to an experiment, which claims to measure a prediction about the temperature dependence made by the Holstein model, with mixed results. This phenomenon was actually among the primary results of T.M. Holstein when he introduced the model[20] now named after him.

Chapter 4 discusses the competition between polaron formation and Cooper pairing, in other words, the possibility of superconductivity. The adiabatic limit is shown to be incompatible with superconductivity, whereas in the anti–adiabatic limit the traditional BCS–treatment does predict it. A method is developed to estimate the region of crossover between these two extremes.

The numerical results presented in chapters 2, 3 and 4 are meant mainly to illustrate methods and mathematics, rather than to say anything about the physics of the model. Chapter 5 will make up for this with a discussion of the phase diagram of the Holstein model in one and two dimensions, and a comparison to the literature. It concludes with a summary of the questions answered and those raised by this thesis.