

Thermal expansion from stochastic nonlinear acoustic fields

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The methods of stochastic mechanics are applied to nonlinear acoustic fields generated by random acoustic radiation sources associated with oscillating lattice sites in nonlinear lattices. The assumption of stochastically independent zero-point and temperature-dependent acoustic fields leads to expressions of the thermodynamic internal and Helmholtz free energies per unit mass in terms of modal energies per unit mass that account for the nonlinearity of the propagation modes. The thermodynamic state functions canonically transform to the familiar results of a system of quantized, simple harmonic oscillators in the linear field limit. The thermal expansion coefficient, derived from the Helmholtz free energy per unit mass, is obtained as a sum of the zero-point modal nonlinearity parameters, weighted by temperature-dependent, modal heat capacities. The relationship is fully anharmonic, in contrast to commonly used quasi-harmonic models, and predicts with excellent agreement the experimentally observed null thermal expansion of vitreous silica at the ‘cross-over’ temperature corresponding to the balance between long wavelength modes that contribute negative nonlinearity parameters to the modal sum and short wavelength modes that contribute positive nonlinearity parameters.

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I. INTRODUCTION

We have shown experimentally [1] the existence of modal radiation stresses and radiation-induced static strains associated with the propagation of coherent, acoustic waves in nonlinear lattices. The acoustic radiation stresses are predicted from the Boltzmann-Ehrenfest principle of adiabatic invariance when the principle is applied to wave propagation in nonlinear, anisotropic, elastic continua [2]. The static strains result from an application of the virial theorem to such media [3]. The statistical nature of these energy principles suggests that the nonlinearity of the propagation medium may be exploited directly to provide a phenomenological link between mechanical radiation and the thermodynamic properties of solids without having to resort to commonly used quasi-linear or quasi-harmonic assumptions.

We consider here the implications of the acoustic radiation stresses to the thermal properties of solids by assuming that the vibrating particles of the lattice are generators of a stochastic, temperature-dependent, nonlinear acoustic radiation field. This notion, together with the assumption of an independent, zero-point acoustic radiation field analogous to that of stochastic electrodynamics [4-7], leads to expressions of the internal and Helmholtz free energies per unit mass in terms of the same acoustical parameters of the material that describe the acoustic radiation stresses and radiation-induced static strains. An expression for the coefficient of thermal expansion follows directly from the Helmholtz free energy density. The expressions for the internal energy and the Helmholtz free energy canonically transform in the linear field limit to the familiar equations obtained from quantum mechanics.

We begin in Section II with derivations of the thermodynamic state functions from classical stochastic nonlinear lattice fields after deriving the relation between the temperature-dependent nonlinear radiation field and the zero-point nonlinear radiation field. The connection of the derived

equations to quantum mechanics is obtained via a canonical transformation of the mode equations in the linear field limit. In Section III an expression for the thermal expansion coefficient is obtained from the Helmholtz free energy in terms of a weighted sum of modal nonlinearity parameters that quantify modal acoustic radiation stresses and radiation-induced static strains in nonlinear lattices [1-3]. Validation of the model is obtained in Section IV from a quantitative assessment of the cross-over temperature at which the thermal expansion of vitreous silica is experimentally measured to be zero.

II. DERIVATION OF THERMODYNAMIC STATE FUNCTIONS FROM STOCHASTIC NONLINEAR ACOUSTIC FIELDS

Studies by Boyer [4-6] of classical, stochastic, linear electromagnetic fields have led to a classical derivation of Planck's blackbody spectrum. An axiomatic approach by Theimer [7], based on Boyer's analysis, leads to the same equations. The similarities in the mathematical descriptions of linear electromagnetic and acoustic fields suggest that assumptions analogous to that of Theimer for electromagnetic fields would also hold for acoustic fields. In order to include the effects of nonlinearity, we consider the fields in question to be time-averaged. Time-averaging not only simplifies the field equations sufficiently that Theimer's mathematical approach can be used, but also provides a direct link with experimentally validated acoustic radiation stresses and radiation-induced static strains.

We thus propose in analogy to the assumptions of Theimer [7] that: (i) there exists a classical, fluctuating, nonlinear acoustic radiation field in a solid at the absolute zero of temperature having an energy per unit mass E_0 ; (ii) the zero-point radiation fluctuates randomly as if it were produced by a large number of incoherent radiation sources associated with the vibrating

lattice particles; (iii) at a finite temperature T the total energy per unit mass E is composed of the zero-point field E_0 and a stochastically, independent, randomly fluctuating, temperature-dependent field E_T such that $E = E_0 + E_T$; (iv) thermal equilibrium is established separately for each acoustic mode ε to first order in the nonlinearity; and (v) the temperature-dependent field E_T^ε for each mode ε obeys well-established temperature-dependent fluctuation theory of classical statistical mechanics.

Assumption (iv) is made plausible by the numerical studies of nonlinear lattices by Fermi, Pasta, and Ulam (FPU) [8], showing that the nonlinearity of interaction between lattice points does not produce an equipartition of energy among the independent linear lattice modes, as expected. Instead, little equipartition occurred and the system periodically returned to its initial state. The FPU studies and others that followed [9] have established ‘nonlinear lattice modes’ as independent dynamical entities. Generally, nonlinear acoustic systems involve the pumping of energy from the fundamental to harmonically-generated oscillations [1-3,9,10] and the nonlinear interaction (mixing) of different lattice modes having different frequencies leads to the generation of other modes with frequencies corresponding to the sum and difference frequencies of the original interacting modes [11-13]. For a system consisting of a very large number of interacting modes in equilibrium, it is assumed that the energy lost from a given mode by energy pumping to other modes is on average compensated by the energy gained by that mode via pumping from the other modes such that the average energy corresponding to a given mode remains constant. In order to consider the total energy associated with a given nonlinear mode without having to consider the details of the energy pumping, we invoke a time-averaging of the dynamical variables such that the mean stress and mean strain for a given mode remain constant at a given temperature.

A. Relationship between zero-point and temperature-dependent fields

The derivation of the relationship between thermodynamic state functions and stochastic nonlinear acoustic fields is dependent on establishing the relationship between the zero-point energy field and the temperature-dependent energy field. It is useful to begin with a consideration of crystalline solids. The nonlinear elastic equations of motion in Lagrangian coordinates embedded in Cartesian space for each mode of a crystalline solid may be transformed into the form [2,14]

$$\frac{\partial^2 P_\varepsilon}{\partial t^2} = s_\varepsilon^2 \left(1 - \beta^\varepsilon \frac{\partial P_\varepsilon}{\partial a} \right) \frac{\partial^2 P_\varepsilon}{\partial a^2} \quad (1)$$

where P_ε is the particle displacement, $\varepsilon = (p, \mathbf{k})$ is a mode index representing a wave of polarization p ($= 1, 2, 3$) corresponding to one quasi-longitudinal mode (denoted by '1') and two quasi-transverse modes (denoted by '2' and '3') and the wave propagation vector \mathbf{k} , s_ε is the infinitesimal amplitude wave phase velocity for mode ε , $\beta^\varepsilon = \beta^{(p,k)}$ is the modal acoustic (material) nonlinearity parameter, t is time, ' a ' is the transformed Lagrangian coordinate along the direction of wave propagation, and $(\partial P_\varepsilon / \partial a)$ is the displacement gradient for mode ε . The Cartesian components of the unit vector in the direction of wave polarization are given as U_i^p ($i = 1, 2, 3$) for each type of polarization 1, 2, or 3. The Cartesian components N_i of the unit vector N in the direction of wave propagation are given as $N_i = k_i / |\mathbf{k}|$ ($i = 1, 2, 3$). The Earnshaw particle velocity $(\partial P_\varepsilon / \partial t)$ solution to Eq.(1) may be written as [15]

$$\frac{\partial P_\varepsilon}{\partial t} = \xi \sin(\omega_\varepsilon t + \theta_\varepsilon) \quad (2)$$

where

$$\theta_\varepsilon = k_\varepsilon a \left(\frac{\beta^\varepsilon}{2s_\varepsilon} \frac{\partial P_\varepsilon}{\partial t} - 1 \right) + \chi_\varepsilon, \quad (3)$$

where $k_\varepsilon = |\mathbf{k}_\varepsilon| = \omega_\varepsilon / s_\varepsilon = 2\pi / \lambda_\varepsilon$ (λ_ε is wavelength) and χ_ε is an arbitrary phase term for mode ε . The modal nonlinearity parameters $\beta^\varepsilon = \beta^{(p, \mathbf{k})}$ are obtained, assuming no initial stresses, as [14]

$$\beta^\varepsilon = \beta^{(p, \mathbf{k})} = - \frac{\sum_{ijklmn} (C_{jlmn} \delta_{ik} + C_{ijnl} \delta_{km} + C_{jknl} \delta_{im} + C_{ijklmn}) N_j N_l N_n U_i^p U_k^p U_m^p}{\sum_{ijkl} C_{ijkl} N_j N_l U_i^p U_k^p} \quad (4)$$

where δ_{ij} are Kronecker deltas, $N = \mathbf{k} / |\mathbf{k}|$, and C_{ijklmn} and C_{ijklmn} , respectively, are the second-order and third-order Brugger elastic constants.

Consider first the zero-point acoustic radiation field. We assume a large number N_0 of incoherent radiation sources in the crystal associated with the lattice points. We assume that for a given mode ε each of the incoherent radiation sources contributes to the zero-point modal particle velocity field $(\partial P_\varepsilon / \partial t)_o$ a component $(\partial P_\varepsilon / \partial t)_{o,r}$ ($r = 1, 2, 3, \dots, N_0$) with effectively random amplitudes $\xi_{o,r}$ and phases $\theta_{\varepsilon,r}$ at a given point in the crystal. The subscripted 'o' denotes zero-point field. The zero-point particle velocity field $(\partial P_\varepsilon / \partial t)_o$ for mode ε is given by the sum over all sources contributing to mode ε as

$$\left(\frac{\partial P_\varepsilon}{\partial t} \right)_o = \sum_{r=1}^{N_0} \xi_{o,r} \sin(\omega_\varepsilon t + \theta_{\varepsilon,r}). \quad (5)$$

The zero-point energy per unit mass E_0^ε for mode ε may be written as [2]

$$E_0^\varepsilon = \frac{1}{2} \left(\frac{\partial P_\varepsilon}{\partial t} \right)_0^2 + \frac{1}{2} s_\varepsilon^2 \left(\frac{\partial P_\varepsilon}{\partial a} \right)_0^2 - \frac{1}{6} \beta^\varepsilon s_\varepsilon^2 \left(\frac{\partial P_\varepsilon}{\partial a} \right)_0^3 \approx \left(\frac{\partial P_\varepsilon}{\partial t} \right)_0^2 - \frac{1}{12} \frac{\beta^\varepsilon}{s_\varepsilon} \left(\frac{\partial P_\varepsilon}{\partial t} \right)_0^3 \quad (6)$$

to first order in the nonlinearity. The last equality results from the compatibility condition [2] $(\partial P_\varepsilon / \partial a) = -s_\varepsilon^{-1} (\partial P_\varepsilon / \partial t) + (\beta_\varepsilon / 4s_\varepsilon^2) (\partial P_\varepsilon / \partial t)^2$. It is noted from Eq.(5) that the zero-point particle velocity field $(\partial P_\varepsilon / \partial t)_0$ for mode ε is dependent on both time and phase. The average field is thus obtained by averaging both time and phase. Substituting Eq.(5) in Eq.(6) and time-averaging the resulting expression, we obtain

$$\overline{E_0^\varepsilon} = \frac{1}{2} \sum_{r,s} \xi_{0,r} \xi_{0,s} \cos(\theta_{\varepsilon,r} - \theta_{\varepsilon,s}) \quad (7)$$

where the overbar denotes time-average. When averaging over random phases, $\cos(\theta_{\varepsilon,r} - \theta_{\varepsilon,s}) = \delta_{rs}$, where δ_{rs} are the Kronecker deltas, and Eq.(7) reduces to

$$\langle \overline{E_0^\varepsilon} \rangle = \frac{1}{2} \sum_s \xi_{0,s}^2 \quad (8)$$

where the angular brackets denote phase-average. We similarly find

$$\langle (\overline{E_0^\varepsilon})^2 \rangle = \frac{1}{4} (\sum_{r,t} \xi_{0,r}^2 \xi_{0,t}^2 + \sum_{s,u} \xi_{0,s}^2 \xi_{0,u}^2) = 2 \langle \overline{E_0^\varepsilon} \rangle^2 \quad (9)$$

where the last equality follows from Eq.(8). From Eq.(9) the mean-square fluctuation of the time-phase-averaged zero-point energy field for mode ε is found to be

$$\langle (\delta \overline{E}_o^\varepsilon)^2 \rangle = \langle (\overline{E}_o^\varepsilon)^2 \rangle - \langle \overline{E}_o^\varepsilon \rangle^2 = \langle \overline{E}_o^\varepsilon \rangle^2 . \quad (10)$$

In accordance with assumption (iii) given in Section II, we now add to the zero-point field a temperature-dependent field corresponding to mode ε that is produced by N_T random radiation sources having corresponding modal particle velocity amplitudes $(\partial P_\varepsilon / \partial t)_{T,\sigma}$ ($\sigma = 1, 2, 3, \dots, N_T$). The total fluctuating particle velocity field $(\partial P_\varepsilon / \partial t)$ at a given point in the material for each mode ε is then given as

$$\frac{\partial P_\varepsilon}{\partial t} = \left(\frac{\partial P_\varepsilon}{\partial t} \right)_o + \left(\frac{\partial P_\varepsilon}{\partial t} \right)_T = \sum_{s=1}^{N_o} \xi_{o,s} \sin(\omega_\varepsilon t + \theta_{\varepsilon,s}) + \sum_{\sigma=1}^{N_T} \xi_{T,\sigma} \sin(\omega_\varepsilon t + \theta_{\varepsilon,\sigma}) \quad (11)$$

where the subscripted T denotes a stochastically independent, temperature-dependent field contribution to mode ε . The total time-averaged energy per unit mass for mode ε is obtained from Eq.(11) as

$$\begin{aligned} \overline{E}^\varepsilon = \overline{E}_o^\varepsilon + \overline{E}_T^\varepsilon = & \frac{1}{2} \sum_{r,s} \xi_{o,r} \xi_{o,s} \cos(\theta_{\varepsilon,r} - \theta_{\varepsilon,s}) \\ & + \left[\frac{1}{2} \sum_{\sigma,\tau} \xi_{T,\sigma} \xi_{T,\tau} \cos(\theta_{\varepsilon,\sigma} - \theta_{\varepsilon,\tau}) + \sum_{r,\sigma} \xi_{o,r} \xi_{T,\sigma} \cos(\theta_{\varepsilon,r} - \theta_{\varepsilon,\sigma}) \right] \end{aligned} \quad (12)$$

where the first term in the right equality of Eq.(12) is the time-averaged zero-point energy per unit mass $\overline{E}_o^\varepsilon$ and the terms within the square brackets comprise the time-averaged temperature-dependent energy per unit mass $\overline{E}_T^\varepsilon$. Phase-averaging Eq.(12) leads to

$$\langle \overline{E}^\varepsilon \rangle = \langle \overline{E}_o^\varepsilon \rangle + \langle \overline{E}_T^\varepsilon \rangle , \quad (13)$$

$$\langle (\overline{E^\varepsilon})^2 \rangle = 2\langle \overline{E^\varepsilon} \rangle^2 , \quad (14)$$

and

$$\langle (\delta \overline{E^\varepsilon})^2 \rangle = \langle (\overline{E^\varepsilon})^2 \rangle - \langle \overline{E^\varepsilon} \rangle^2 = \langle \overline{E^\varepsilon} \rangle^2 . \quad (15)$$

We now re-write Eq.(15) in the form

$$\begin{aligned} \langle (\delta \overline{E^\varepsilon})^2 \rangle &= \langle (\overline{E_0^\varepsilon} + \overline{E_T^\varepsilon})^2 \rangle - (\langle \overline{E_0^\varepsilon} \rangle + \langle \overline{E_T^\varepsilon} \rangle)^2 \\ &= \langle (\delta \overline{E_0^\varepsilon})^2 \rangle + \langle (\delta \overline{E_T^\varepsilon})^2 \rangle \end{aligned} \quad (16)$$

where the last equality follows from the stochastic independence of the zero-point and temperature-dependent fields

$$\langle \overline{E_0^\varepsilon} \overline{E_T^\varepsilon} \rangle = \langle \overline{E_0^\varepsilon} \rangle \langle \overline{E_T^\varepsilon} \rangle . \quad (17)$$

It is straightforward to verify Eq.(17) from Eq.(12). Since $\langle \overline{E_0^\varepsilon} \rangle$ are time and phase averaged modal energies per unit mass, we define $\langle \overline{E_{0V}^\varepsilon} \rangle = \rho_0 V \langle \overline{E_0^\varepsilon} \rangle$ as modal energies associated with a volume V of material having a mass density ρ_0 . In accordance with assumption (v) given in Section II, we now impose that the temperature-dependent modal contribution E_T^ε , hence E_{TV}^ε , obeys the temperature-dependent fluctuation theory of classical statistical mechanics. We thus write the mean-square modal fluctuations $\langle (\delta \overline{E_T^\varepsilon})^2 \rangle$ of $\overline{E_T^\varepsilon}$ as [16]

$$\langle (\delta \overline{E}_T^\varepsilon)^2 \rangle = \langle (\overline{E}_T^\varepsilon)^2 \rangle - \langle \overline{E}_T^\varepsilon \rangle^2 = \frac{k_B T^2}{\rho_0 V} \frac{\partial \langle \overline{E}_T^\varepsilon \rangle}{\partial T} \quad (18)$$

where k_B is Boltzmann's constant and T is temperature. From Eqs.(10), (13), (15), and (16) we re-write Eq.(18) as

$$\langle \overline{E}_T^\varepsilon \rangle^2 + 2 \langle \overline{E}_0^\varepsilon \rangle \langle \overline{E}_T^\varepsilon \rangle = \frac{k_B T^2}{\rho_0 V} \frac{\partial \langle \overline{E}_T^\varepsilon \rangle}{\partial T} \quad (19)$$

with solution

$$\langle \overline{E}_T^\varepsilon \rangle = \frac{2 \langle \overline{E}_0^\varepsilon \rangle}{e^{2\rho_0 V \langle \overline{E}_0^\varepsilon \rangle / k_B T} - 1} . \quad (20)$$

Eq.(20) gives the temperature-dependent, time and phase-averaged energy per unit mass as a function of the zero-point, time and phase-averaged energy per unit mass. The equation is remarkably similar to the Planck distribution equation. We now consider the relationship of Eq.(20) to the Planck equation and the thermodynamic state functions.

B. Thermodynamic state functions

In the linear field limit the zero-point modal nonlinearity parameters β_0^ε are zero. Hence, the zero-point modal Hamiltonian density H_0^ε may be written as

$$H_0^\varepsilon = (2\rho_0)^{-1} \pi_\varepsilon^2 + \frac{1}{2} \rho_0 s_\varepsilon^2 \left(\frac{\partial p_\varepsilon}{\partial a} \right)^2 \quad (21)$$

where π_ε is the momentum density for mode ε , ρ_0 is the mass density of the solid, and s_ε is the elastic wave phase velocity for mode ε . We impose on Eq.(21) the Bloch condition for periodic

lattices, $P_\varepsilon = \eta_\varepsilon(t)\exp(ik_\varepsilon a)$, where $k_\varepsilon = \omega_\varepsilon/s_\varepsilon$ and $\eta_\varepsilon(t)$ is a time-dependent, periodic function.

Imposing the Bloch condition in Eq.(21) leads to a modal Hamiltonian density in the mathematical form of a simple harmonic oscillator

$$H_0^\varepsilon = (2\rho_0)^{-1}\pi_\varepsilon^2 + \frac{1}{2}\rho_0\omega_\varepsilon^2\eta_\varepsilon^2 \quad . \quad (22)$$

The link to the Planck distribution function is obtained by imposing on Eq.(22) the canonical transformation

$$\pi_\varepsilon = (2\rho_0\omega_\varepsilon\Pi_\varepsilon)^{1/2} \cos Q_\varepsilon \quad , \quad \eta_\varepsilon = \left(\frac{2\Pi_\varepsilon}{\rho_0\omega_\varepsilon}\right)^{1/2} \sin Q_\varepsilon \quad (23)$$

that results from the generating function $G(\eta_\varepsilon, Q_\varepsilon) = (1/2)\rho_0\omega_\varepsilon\eta_\varepsilon^2 \cot Q_\varepsilon$, where Q_ε is the transformed generalized coordinate and Π_ε is the transformed conjugate momentum density [17].

Eqs.(23) canonically transform Eq.(22) into $H_0^\varepsilon = \Pi_\varepsilon\omega_\varepsilon$. Since H_0^ε is independent of Q_ε (i.e., cyclic with respect to Q_ε), Π_ε is a constant of the motion that we choose to write as $\hbar/2V$. From the total energy relation for a conservative system $H_0^\varepsilon = \rho_0 E_0^\varepsilon$, we obtain $E_0^\varepsilon = \hbar\omega_\varepsilon/2\rho_0V$ and identify E_0^ε as $E_0^\varepsilon = \overline{E_0^\varepsilon}$. Substituting this expression in Eq.(20), we obtain the Planck distribution formula

$$\overline{E_{TV}^\varepsilon} = \rho_0 V \overline{E_T^\varepsilon} = \frac{\hbar\omega_\varepsilon}{e^{\hbar\omega_\varepsilon/k_B T} - 1} \quad (24)$$

where we identify \hbar as the angular Planck constant.

We now return to the case where the modal nonlinearity parameters β^ε are nonzero. We consider that the internal energy per unit mass U of the crystal is the sum over all modes of the zero-point and temperature-dependent modal energies per unit mass in accordance with the assumptions (iii) and (iv) given above, plus a configurational energy ϕ corresponding to the potential energy per unit mass when the lattice particles are at rest in their mean positions. Using Eq.(20), we thus write the internal energy per unit mass as

$$U = \phi + \sum_\varepsilon \langle \overline{E}_0^\varepsilon \rangle \left[1 + 2 \left(e^{2\rho_0 V \langle \overline{E}_0^\varepsilon \rangle / k_B T} - 1 \right)^{-1} \right] . \quad (25)$$

The internal energy per unit mass U and the Helmholtz free energy per unit mass F are related by the Gibbs-Helmholtz equation

$$U = -T^2 \frac{\partial}{\partial T} \left(\frac{F}{T} \right) . \quad (26)$$

From Eqs.(25) and (26) we obtain the Helmholtz free energy per unit mass as

$$F = \phi + \sum_\varepsilon \left[\langle \overline{E}_0^\varepsilon \rangle + \frac{k_B T}{\rho_0 V} \ln \left(1 - e^{-2\rho_0 V \langle \overline{E}_0^\varepsilon \rangle / k_B T} \right) \right] . \quad (27)$$

In the linear field limit, where $\langle \overline{E}_0^\varepsilon \rangle$ canonically transforms to $\hbar \omega_\varepsilon / 2\rho_0 V$, the internal energy per unit mass and the Helmholtz free energy per unit mass transforms to that obtained from quantum mechanics.

III. RELATIONSHIP BETWEEN THERMAL EXPANSION AND MATERIAL NONLINEARITY PARAMETERS

The thermal expansion tensor α_{jk} is obtained from the Helmholtz free energy density $\rho_0 F$ as [18,19]

$$\alpha_{jk} = -\sum_{r,s} S_{jkrs}^T \rho_0 \left(\frac{\partial^2 F}{\partial \eta_{rs} \partial T} \right)_{\eta=0} \quad (28)$$

where S_{jkrs}^T are the isothermal compliance coefficients, and η_{rs} are the Lagrangian strains. It is noted that $\rho_0 (\partial F / \partial \eta_{rs}) = t_{rs}$ corresponds to the second Piola-Kirchhoff stresses (thermodynamic tensions) t_{rs} [20]. Since we are considering that the thermal expansion results from a stochastic nonlinear acoustic field generated by a large number of acoustic sources, it is appropriate to consider F as a function of the displacement gradients u_{rs} such that the stresses are the Lagrangian stresses $\sigma_{rs} = \rho_0 (\partial F / \partial u_{rs})$ relevant to acoustic wave propagation [2,14,20]. In terms of the Lagrangian stresses, the thermal expansion tensor is given as

$$\alpha_{jk} = -\sum_{r,s} S_{jkrs}^T \rho_0 \left(\frac{\partial^2 F}{\partial u_{rs} \partial T} \right)_{u=0} = -\sum_{r,s} S_{jkrs}^T \left(\frac{\partial \sigma_{rs}}{\partial T} \right)_{u=0}. \quad (29)$$

The derivatives with respect to the displacement gradients can be expressed in terms of derivatives with respect to the modal displacement gradients $(\partial P_\varepsilon / \partial a)$ in the transformed Lagrangian coordinate ‘ a ’ along the direction of wave propagation as [14]

$$\frac{\partial}{\partial u_{rs}} = U_r^p N_s \frac{\partial}{\partial (\partial P_\varepsilon / \partial a)} \quad (30)$$

where $N_s = k_s/|\mathbf{k}|$ ($s = 1, 2, 3$) are the Cartesian components of the unit vector N along the direction of wave propagation and U_r^p ($r = 1, 2, 3$) are the Cartesian components of the wave polarization vector corresponding to polarization p ($p = 1, 2, 3$). From Eqs.(27), (29) and (30), we obtain

$$\alpha_{jk} = -\sum_{\varepsilon} \sum_{rs} S_{jkr}^T C^{\varepsilon} g_{rs}^{\varepsilon} \quad (31)$$

where

$$C^{\varepsilon} = C^{(p,\mathbf{k})}(T) = 4\rho_0^2 V \frac{\langle \overline{E_0^{\varepsilon}} \rangle^2}{k_B T^2} e^{2\rho_0 V \langle \overline{E_0^{\varepsilon}} \rangle / k_B T} \left(e^{2\rho_0 V \langle \overline{E_0^{\varepsilon}} \rangle / k_B T} - 1 \right)^{-2} \quad (32)$$

are the modal specific heat capacities at temperature T and, from the first equality in Eq.(6), we define the fractional variation in the mean energy per unit mass as

$$g_{rs}^{\varepsilon} = \frac{1}{\langle \overline{E_0^{\varepsilon}} \rangle} \left\langle \frac{\partial \overline{E_0^{\varepsilon}}}{\partial u_{rs}} \right\rangle = -\frac{1}{4} U_r^p N_s \beta_0^{\varepsilon} = -G_{rs}^{\varepsilon} \beta_0^{\varepsilon} \quad (33)$$

where $\varepsilon = (p,\mathbf{k})$, $G_{rs}^{\varepsilon} = U_r^p N_s$, and β_0^{ε} is the zero-point modal nonlinearity parameter. The last two equalities in Eq.(33) result from Eq.(30) and the compatibility condition [2] $(\partial P_{\varepsilon} / \partial a) = -s_{\varepsilon}^{-1} (\partial P_{\varepsilon} / \partial t) + (\beta_{\varepsilon} / 4s_{\varepsilon}^2) (\partial P_{\varepsilon} / \partial t)^2$.

The volume expansion coefficient α is obtained from the trace of Eq.(31) as

$$\alpha = \sum_{\varepsilon} \sum_{jrs} S_{jjrs}^T C^{\varepsilon} G_{rs}^{\varepsilon} \beta_0^{\varepsilon} = \sum_{p,\mathbf{k}} \sum_{jrs} S_{jjrs}^T C^{(p,\mathbf{k})} G_{rs}^{(p,\mathbf{k})} \beta_0^{(p,\mathbf{k})} . \quad (34)$$

For cubic crystals, the number of independent compliance coefficients is restricted to three such that $r = s$. This restriction permits Eq.(34) to be written as

$$\alpha = K_T \sum_{\varepsilon} \sum_m C^{\varepsilon} G_{mm}^{\varepsilon} \beta_o^{\varepsilon} = \frac{K_T}{4} \sum_{p,k} C^{(p,k)} G_{rs}^{(p,k)} \beta_o^{(p,k)} \quad (35)$$

where $K_T = \sum_{j,r} S_{jjrr}^T$ is the compressibility.

The thermal expansion coefficient given by Eq.(35) is expressed as a discrete sum over all lattice modes. Since the wave vectors \mathbf{k} are continuous functions of direction and frequency, it is appropriate to re-write the discrete sum as an integral over the density of states in reciprocal space such that $\sum_k \rightarrow \int d\mathbf{k} = (V/8\pi^3) \iiint d^3k$. The thermal expansion coefficient can be calculated from Eq.(35) via the Debye approach using the following assumptions [18,19]: (i) Only acoustic modes ($p = 1, 2, 3$) are considered (optical modes can be added as appropriate for non-monatomic materials), (ii) the acoustic modes obey the Debye distribution function $\mathcal{D}_o^{\varepsilon}(p,\mathbf{k}) \sim k^2$ within the Debye sphere of the reciprocal lattice for each polarization p , (iii) the maximum value of k along any direction equals the Debye radius $k_D = (6\pi^2/V)^{1/3}$ where V is the volume of the primitive cell, and (iv) the acoustic modes obey the Born-von Karman dispersion relationship

$$\omega_{\varepsilon} = \frac{2}{\pi} \omega_D \sin\left(\frac{\pi k_{\varepsilon}}{2 k_D}\right) = \frac{2}{\pi} s_{\varepsilon} k_D \sin\left(\frac{\pi k_{\varepsilon}}{2 k_D}\right) \quad (36)$$

where the last equality follows from the relation $\omega_D = s_{\varepsilon} k_D$ with s_{ε} being the modal phase velocity.

The above assumptions provide that for a given wave propagating in the unit direction $\mathbf{N} = \mathbf{k}/k$, $k = |\mathbf{k}|$ ranges from zero to k_D for each mode. Since \mathbf{k} remains within the Debye sphere of radius k_D , the summation over \mathbf{k} can be replaced by an integral over the volume of the Debye sphere as

$$\Sigma_k \rightarrow \left(\frac{V}{8\pi^3}\right) \int_0^{k_D} \int_{\Omega} k^2 dk d\Omega \quad (37)$$

where Ω is the solid angle. The factor $(V/8\pi^3)$ is the density of states within the Debye sphere.

Applying Eq.(37) to Eq.(35) , we obtain

$$\alpha = \frac{K_T}{4} \sum_p \sum_m \int_{\Omega} d\Omega C^{(p,k)} U_m^p N_m \beta_o^{(p,k)} \quad (38)$$

where the modal specific heat capacities $C^{(p,k)}$ are given by

$$C^{(p,k)}(T) = 9Nk_B \left(\frac{T}{\Theta_D}\right)^3 \int_0^{\Theta_D/T} \frac{z^4 e^z}{(e^z - 1)^2} dz \quad (39)$$

where

$$z = \left(\frac{\hbar\omega}{k_B T}\right), \quad (40)$$

N is the number of atoms in the volume of material and the Debye temperature Θ_D is defined as

$$\Theta_D = \left(\frac{\hbar\omega_D}{k_B}\right) \quad . \quad (41)$$

For expediency in obtaining the modal heat capacities of Eqs.(39), we have used the results obtained in the linear field limit, where $\langle \overline{E_o^\varepsilon} \rangle$ canonically transforms to $\hbar\omega_\varepsilon/2\rho_o V$ and ω is related to k via Eq.(36). The approximation is justified on the grounds that the time average of the cubic term on the right in Eq.(6) is zero for sinusoidal oscillations. However, the time average of the

strain derivative of the cubic term in Eq.(6) is nonzero for sinusoidal oscillations and is given by Eq.(32).

The Debye temperature θ_D is calculated from measurements of the modal sound velocities $s_{(p,N)}$ as [18,19]

$$\frac{1}{\theta_D^3} = \frac{3\pi^2}{4} \left(\frac{k_B}{\hbar k_D} \right)^3 \sum_p \int_{\Omega} \frac{d\Omega}{s_{(p,N)}^3} \quad (42)$$

For each chosen wave propagation direction N in the Debye sphere, the modal wave speed $s_{(p,N)}$ and the modal wave polarization vector U are obtained from the Christoffel expression [18,19]

$$\sum_k [\sum_{jl} C_{ijkl} N_j N_l - \rho_0 s_{(p,N)}^2 \delta_{ik}] U_k^p = 0 \quad (43)$$

where C_{ijkl} are the second-order Brugger elastic constants.

IV. ASSESSMENT OF THERMAL EXPANSION CROSS-OVER TEMPERATURE FOR VITREOUS SILICA

A test of the validity of the above model is provided by thermal expansion measurements of vitreous silica. The thermal expansion of vitreous silica is experimentally observed to be zero at a temperature, which we shall call the ‘cross-over’ temperature. Below the cross-over temperature the thermal expansion is negative while above that temperature the thermal expansion is positive. The structure of vitreous silica provides a clue for the cross-over behavior. The macroscale structure of vitreous silica is amorphous but assumes a ‘quartz-like’ (i.e., crystal-like) nanoscale structure within a radius of several atomic diameters around any given material point as the result of strong, covalent, tetrahedral Si-O-Si bonds [21,22].

At low temperatures long wavelength acoustic modes, associated with the amorphous structure, dominate the thermal expansion. In this regime the lattice appears random and the acoustic modes behave as if propagating in an isotropic lattice where the ‘lattice periodicity’ corresponds to that of the acoustic wavelength. The isotropic lattice leads to non-zero values of the longitudinal nonlinearity parameters [23]. However, the transverse (shear) nonlinearity parameters for isotropic lattices are zero [23], which means that no shear acoustic radiation stresses (mean acoustic shear stresses) or shear radiation-induced static strains (mean acoustic shear strains) are generated. For vitreous silica the nonlinearity parameters corresponding to the longitudinal modes are negative [24] and result in a negative thermal expansion coefficient at low temperatures where the long wavelength modes dominate the frequency spectrum.

At high temperatures the short wavelength modes, associated with the local quartz-like structure, become more populated and increasingly dominate the thermal expansion as the temperature increases. The nonlinearity parameters corresponding to these short wavelength modes, calculated from Eq.(4) using measured values of the second and third-order elastic constants [25], are dominantly positive and result in a positive contribution to the thermal expansion coefficient. The present model provides a means of understanding the cross-over temperature as the temperature corresponding to balance between long wavelength modes that contribute negative nonlinearity parameters to the modal sum in Eqs.(34) and (35), and short wavelength modes that contribute positive nonlinearity parameters.

It is assumed that the contributions to the thermal expansion coefficient from the wavelength-dependent amorphous and quartz-like (crystal-like) structures of vitreous silica may be expressed as

$$\alpha = \sum_{\varepsilon} \sum_{jrs} S_{jjrs}^T C^{\varepsilon} G_{rs}^{\varepsilon} \beta_0^{\varepsilon} = \alpha^{amorph} + \alpha^{cryst} \quad (44)$$

where

$$\alpha^{amorph} = K_T^{amorph} \sum_{\varepsilon(k < k_c)} \sum_m C^\varepsilon G_{mm}^\varepsilon (\beta_o^\varepsilon)^{amorph} \approx K_T^{amorph} C^{amorph} \beta_o^{amorph} \quad (45)$$

is the amorphous contribution to the thermal expansion coefficient α corresponding to long wavelength modes. In this regime $k < k_c$ (i.e., $\lambda > \lambda_c$, λ is wavelength) where k_c is the critical wavenumber below which the wavelength is assumed be too long to respond to the quartz-like structure. The assumption is based on the assessed atomic pair correlation functions for vitreous silica [26], where the crystal-like structure appears to fade into an amorphous structure beyond a range of five or six atomic diameters from an arbitrary fixed point in the solid. It is assumed in the last equality in Eq.(45) that the long wavelength modal nonlinearity parameters in the isotropic structure has the same value $(\beta_o^\varepsilon)^{amorph} = \beta_o^{amorph}$ for each mode ε .

The contribution α^{cryst} in Eq.(44) to the thermal expansion coefficient from the quartz-like structures of vitreous silica may be expressed as

$$\alpha^{cryst} = \sum_{\varepsilon(k > k_c)} \sum_{jrs} S_{jjrs}^T C^\varepsilon G_{rs}^\varepsilon (\beta_o^\varepsilon)^{cryst} \quad (46)$$

where $k > k_c$. Since the quartz-like structures within the material occur locally at every point in the material with random orientations, the nonlinearity parameters $(\beta_o^\varepsilon)^{cryst}$ assume an average value for small wavelength propagation similar to that of a quasi-isotropic material. In such case Eq.(46) may be approximated as the contribution from quasi-isotropic, polycrystalline quartz and evaluated as

$$\alpha^{cryst} \rightarrow \alpha^{poly} = K_T^{poly} C^{poly} \beta_o^{poly} \quad (47)$$

where the modal nonlinearity parameters $(\beta_o^\varepsilon)^{poly}$ are assumed to have roughly the same value $(\beta_o^\varepsilon)^{poly} = \beta_o^{poly}$ for each mode ε in the quasi-isotropic, polycrystalline regime.

The Debye temperature for quartz is assessed from thermal measurements as [27] $\Theta_D^{quartz} = 255$ K and that for vitreous silica is assessed from ultrasonic (long wavelength) measurements, corresponding to the amorphous regime, as [28] $\Theta_D^{amorph} = 1340$ K. Thus, for a given temperature T , T/Θ_D^{quartz} is considerably larger than T/Θ_D^{amorph} . This means that the specific heat for the quartz-like contribution is considerably greater than the amorphous contribution at high temperatures. In such case the specific heat can be evaluated from Eq.(38) for large values of T/Θ_D^{quartz} approximately as [18]

$$C^{poly} = 3Nk_B . \quad (48)$$

At low temperatures T/Θ_D^{amorph} is much smaller than T/Θ_D^{quartz} , leading to predominantly amorphous (long wavelength) contributions to the specific heat. In such case T/Θ_D^{amorph} in the upper limit in the integral of Eq.(39) can be approximated as infinite to yield [18]

$$C^{amorph} \approx \frac{12\pi^4}{5} Nk_B \left(\frac{T}{\Theta_D^{amorph}} \right)^3 . \quad (49)$$

It is important to point out that ultrasonic assessments of Θ_D^{amorph} have been found to give a better fit to infrared data of vitreous silica than assessments from thermal data [28]. This indicates that the low-temperature vibrations arise from low-frequency non-acoustic oscillations,

possibly associated with transverse oscillations of the oxygen ion [28]. The non-acoustic oscillations are not captured in the thermal data for the specific heat, but are captured by the ultrasonic measurements of Θ_D^{amorph} . This suggests that the non-acoustic oscillations are an intrinsic feature of the silica structure that is reflected in measurements of the elastic constants and sound velocity. At the cross-over temperature the thermal expansion coefficient, given by Eq.(44), is zero. Thus, assuming $\alpha^{cryst} \rightarrow \alpha^{poly}$, Eqs.(44). (45), and (47) yield

$$K_T^{amorph} C^{amorph} |\beta_o^{amorph}| = K_T^{poly} C^{poly} |\beta_o^{poly}| . \quad (50)$$

Substituting Eqs.(48) and (49) in Eq.(50) leads to an assessment of the cross-over temperature T_{co} as

$$T_{co} = \left(\frac{5}{4\pi^4} \frac{|\beta_o^{poly}|}{|\beta_o^{amorph}|} \frac{K_T^{poly}}{K_T^{amorph}} \right)^{1/3} \Theta_D^{amorph} . \quad (51)$$

The compressibility $K_T^{poly} \approx K_T^{amorph}$ [29,30]. Ultrasonic measurements of the nonlinearity parameter for vitreous silica yield an essentially constant value $\beta^{amorph} = -11.6$ in the range of temperatures 3K – 300K [24,31]. It is assumed that the value holds at the zero-point $T = 0$ as well, such that $\beta_o^{amorph} = -11.6$. The nonlinearity parameter for polycrystalline quartz may be crudely estimated for longitudinal acoustic modes from the averages of the longitudinal mode second and third-order elastic constants. The averaged second-order longitudinal constant is assessed from the data of Heyliger *et al.* [29] as $C_{11} = 96.5$ GPa and the averaged third-order longitudinal constant is assessed from the data of Thurston *et al.* [25] as $C_{111} = -452.3$ GPa, both data sets being measured at roughly 300K. Assuming that the ratio C_{111}/C_{11} is roughly constant

over the range 0K – 300K (as is the case for germanium [32] and copper [33]) we calculate from Eq.(4) the effective longitudinal mode nonlinearity parameter as $\beta_o^{poly} = -[3 + (C_{111}/C_{11})] = 1.7$. Substituting the above values of β_o^{amorph} , β_o^{poly} , K_T^{amorph} , and K_T^{poly} in Eq.(51) yields the cross-over temperature as $T_{co} = 169$ K. This value is in excellent agreement with the measured range of values $\sim (151-188)$ K obtained by White [34] for a variety of vitreous silica samples subjected to various fictive temperatures and annealing history.

V. CONCLUSION

We have applied the methods of stochastic electrodynamics [4-7] to the dynamics of nonlinear lattices by assuming the existence of stochastically independent nonlinear acoustic zero-point and temperature-dependent fields in the material. The assumption leads to an expression of the temperature-dependent field in terms of the zero-point field that is analogous to that of the Planck distribution and to expressions of the thermodynamic internal and Helmholtz free energies in terms of zero-point modal energies associated with the nonlinearity of the propagation medium. Although derived from classical assumptions, the relation between the temperature-dependent and zero-point fields canonically transforms to the Planck equation in the linear field limit and the thermodynamic state functions canonically transform to the familiar results of a system of quantized, simple harmonic oscillators in the linear field limit. The thermal expansion coefficient, derived from the Helmholtz free energy, is obtained as a sum of modal nonlinearity parameters, weighted by temperature-dependent modal heat capacities.

It is important to point out that the nonlinearity parameters are highly ordered according to the crystalline structure and symmetry of the material [14] and serve as scaling parameters for the acoustic radiation stresses and radiation-induced static strains generated for a given mode in solids

[1-3]. Thus, the appearance of β_o^ε in Eqs.(34) and (38) suggests, in consideration of the factor $S_{jkr_s}^T \left(\frac{\partial \sigma_{rs}}{\partial T} \right)_{u=0}$ in Eq.(29) from which Eqs.(34) and (38) are derived, that the thermal expansion coefficient may be viewed as a weighted sum of the temperature derivatives of modal acoustic radiation-induced static strains in the material. In the linear field limit $\beta_o^\varepsilon = 0$ and the present model predicts null values of the thermal expansion coefficient. Such results are expected, since the nonlinearity parameters are measures of the anharmonic properties of the nonlinear lattice. It is noted that the traditional derivation of thermal expansion coefficient is obtained in the linear field limit such that $\beta_o^\varepsilon = 0$ and where, as shown in Section II.B, $\langle \overline{E_o^\varepsilon} \rangle = \hbar \omega_\varepsilon / 2 \rho_o V$. In order to avoid a null value of the thermal expansion for all temperatures in the traditional derivation, the quasi-harmonic postulate is generally imposed whereby the modal vibrational frequencies ω_ε are allowed to be explicit functions of the lattice strains η_{ij} . Thus, the parameters in the quasi-harmonic model analogous to the nonlinearity parameters are the tensor Grüneisen parameters γ_{rs}^ε defined as $\gamma_{rs}^\varepsilon = -(1/\omega_\varepsilon)(\partial \omega_\varepsilon / \partial \eta_{rs})$ [18,19]. It is apparent from Eq.(30) that in the linear field limit the g_{rs}^ε correspond to γ_{rs}^ε .

The dependence of the modal frequencies on strain in the quasi-harmonic model implies that some source external to a given mode produces the strain variations in that mode. The strain variations could occur simply from the interaction with other modes. The nonlinearity parameters, however, provide a direct, quantitative measure of intrinsic modal nonlinearity that does not depend on externally-induced strain variations and are, therefore, more strictly in accordance with the findings of Fermi, Pasta, and Ulam [8] that nonlinear lattice modes occur in dynamical systems as independent dynamical entities. The parameters quantify the distortion of the waveform resulting from self-modulated strains generated by the wave itself along the modal propagation path. The nonlinearity parameters are thus natural measures of material anharmonicity that do not

rely on the ad hoc assumption of quasi-harmonicity to avoid null values of phenomena that are dependent on material anharmonicity. It is thus expected that models of anharmonic phenomena using the fully anharmonic nonlinearity parameters likely lead to results somewhat different from that using the quasi-harmonic Grüneisen parameters. The difference is the subject of further investigation. The excellent agreement between the assessment from the present model of the cross-over temperature of the thermal expansion for vitreous silica and the experimental values provides strong evidence of the validity of the present model.

Finally, the assumption of a classical, stochastically independent, zero-point acoustic radiation field begs some mention of the fundamental implications of such an assumption. The classical electromagnetic analog of the zero-point acoustic field is the zero-point electromagnetic field, which according to Theimer [7] is “in some unknown fashion, equivalent to the ground state of the radiation field in quantum electrodynamics.” Various attempts at understanding the origin of the zero-point electromagnetic radiation have included a consideration of the statistical effects of radiative reaction forces [35] and of the stochastic metric fluctuations in a five-dimensional space-time continuum [36]. It has been suggested [7] that the classical zero-point electromagnetic radiation field may serve as the background radiation postulated by Nelson [37] that allows him to derive Schrödinger’s equation from classical random-walk particle motion. Considerations related to quantum mechanics as an emergent stochastic phenomenon have been advanced by Adler [38], ‘t Hooft [39], and Smolin [40]. The zero-point electromagnetic field is generally acknowledged to be the origin of the Casimir forces [41] and, as such, the zero-point field is manifested as a measurable entity arising from mechanical forces via field-material particle interactions. The present work provides a connection between classical stochastic mechanics and quantum mechanics for nonlinear lattices that leads quite naturally, in the spirit of the Fermi-Pasta-Ulam

numerical studies [8], to the establishment of fully anharmonic lattice modes in describing the thermal properties of solids.

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