



Theory of quantum paraelectrics and the metaelectric transition

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We present a microscopic model of the quantum paraelectric-ferroelectric phase transition with a focus on the influence of coupled fluctuating phonon modes. These may drive the continuous phase-transition first order through a metaelectric transition and furthermore stimulate the emergence of a textured phase that preempts the transition. We discuss two further consequences of fluctuations, first for the heat capacity, and second we show that the inverse paraelectric susceptibility displays $\chi^{-1} \sim T^2$ quantum critical behavior, and can also adopt a characteristic minimum with temperature. Finally, we discuss the observable consequences of our results.

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I. INTRODUCTION

Ferroelectric materials feature in many modern day electronic devices including computer memory and capacitors, and are a simple setting for studying quantum criticality.¹⁻³ In this paper we focus on the family of displacive ferroelectrics where the optical lattice modes condense, forming a structural distortion. Near to quantum criticality excitations can become highly degenerate and new phases can emerge. Motivated by recent experiments that signal the emergence of novel quantum critical behavior in ferroelectrics,¹ we explore the possibility that transverse components of polar fluctuating phonons conspire to drive a first-order displacive metaelectric transition and investigate the implications for the inverse susceptibility.

The soft-mode optical phonons in ferroelectrics can be well described by a bosonic field theory. If the dynamics were not damped by free electrons and the interactions remain short ranged then the general quantum-critical behavior would adhere to the well-established rules reviewed in Ref. 4. However, in ferroelectrics the motion of the atoms in optical modes leads to the emergence of electric dipoles. A good description of these long-range dipole forces is essential to properly describe the ferroelectric transition. The effect of long-range dipolar forces was first studied by Rechester⁵ and Khmel'nitskii and Shneerson.⁶ Aharony and Fisher⁷ found that anisotropies associated with the dipolar interaction led to a universality class in the classical ferroelectric. The quantum ferroelectric phase transition in the mean-field approximation, and its universality class, was studied by Roussev and Millis.⁸ However, recent experimental evidence points to new physics that emerges close to quantum criticality; for example, the coexistence of a quantum paraelectric phase with a quantum ferroelectric phase in ¹⁸O-exchanged SrTiO₃ provides strong evidence for a first-order phase transition.¹ Additional motivation to study ferroelectrics arises from the inverse dielectric constant behavior of SrTiO₃ which falls at low temperature before increasing as $\epsilon^{-1} \sim T^2$ at intermediate temperatures and rises as $\epsilon^{-1} \sim T$ at high temperature. One suggestion is that new phenomena are driven by the coupling of acoustic to optical phonons.^{3,6,9} However, inspired by the ramifications of quantum fluctuations in ferromagnets,¹⁰ we show that the transverse coupling

of fluctuating phonons can drive a first-order metaelectric transition.

Having realized that fluctuations can cause the emergence of a first-order transition it is natural to search for further phase reconstruction. Motivated by the development of a textured Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase^{11,12} and evidence for a textured ferromagnetic state near to the ferromagnetic first-order transition,¹³⁻¹⁵ here we search for the emergence of an analogous textured ferroelectric phase. Finally, to connect to prevailing experimental methods, we derive an appropriate expression for the inverse susceptibility that is consistent with recent experimental results^{3,16,17} over a wide range of the phase diagram and demonstrate that the transverse coupling of fluctuating phonons could cause it to have a characteristic minimum at low temperature.

II. ACTION AND MEAN-FIELD THEORY

We adopt a bosonic field theory to describe the soft optical phonon modes that should recover the main physical behavior of the system. The order parameter of the theory is the local polarization $\phi(\mathbf{x}, t) = \sum_{i=1}^n e_i \mathbf{r}_i(\mathbf{x}, t)$, which is formally defined for one unit cell at \mathbf{x} containing n atoms of charge e_i each individually displaced through \mathbf{r}_i by the optic mode. As the optical phonon softens, the action develops an instability and the order parameter must describe both thermal and quantum fluctuations. Following Ref. 8 we describe the action in three-dimensional space and imaginary time via the Ginzburg-Landau phenomenology

$$S = \int_0^\beta \left\{ \sum_{\mathbf{q}, \alpha, \beta} \left[\left(\frac{a^2}{c^2} \partial_\tau^2 + a^2 q^2 + r + f q_\alpha^2 \right) \delta_{\alpha, \beta} + (g - h q^2) \frac{q_\alpha q_\beta}{q^2} \right] \phi_\alpha(\mathbf{q}) \phi_\beta(-\mathbf{q}) + \sum_{\alpha, \beta, \{\mathbf{q}_i\}} (u + v \delta_{\alpha, \beta}) \phi_\alpha(\mathbf{q}_1) \phi_\alpha(\mathbf{q}_2) \phi_\beta(\mathbf{q}_3) \phi_\beta(\mathbf{q}_4) \right\} d\tau, \quad (1)$$

where a is the lattice constant, c is the speed of the phonons, $q^2 = \sum_\alpha q_\alpha^2$, the dimensionless momenta $-\pi < q_\alpha \leq \pi$, and the second summation is carried out under the conservation of

TABLE I. Model parameters for the ferroelectrics SrTiO₃ and KTaO₃ (Refs. 3, 8, 18, and 19).

	E_0 (meV)	a (Å)	$\hbar c$ (meV)	r	f	g	h
SrTiO ₃	4.47	3.9	5.55	5.31	55.7	0.39	5.1
KTaO ₃	10.6	3.9	13.1	9.77	472	39.2	165

momentum ($\mathbf{q}_1 + \mathbf{q}_2 + \mathbf{q}_3 + \mathbf{q}_4 = 0$). Since the field ϕ describes an electric dipole, the action includes a long-range dipole interaction, and also a coupling to the underlying lattice through the parameters r , f , g , and h . The terms u and v that describe the local anharmonic interactions give a net positive contribution which ensures that the polarization remains bounded. In general these parameters are tensorial but for simplicity we have assumed that they adopt cubic symmetry. Estimates for the parameters shown in Table I were obtained from *ab initio* calculations^{8,18,19} in the two-key ferroelectrics SrTiO₃ and KTaO₃.³ The typical energy scale of ferroelectric fluctuations along (100) is $E_0 = \hbar\pi c/a$; using this definition we can then employ a dimensionless bosonic Matsubara frequency $\tilde{\omega} = \omega/E_0$ and a dimensionless temperature $\tilde{T} = T/E_0$. Throughout the paper we adopt the units $a = \hbar = k_B = 1$.

To establish the connection to previous work we first consider the mean-field phase diagram that is sketched in Fig. 1. Making the ansatz that the ground state is uniform we obtain the action $S = r\phi^2 + (u+v)\phi^4$, where $\phi = |\phi|$. When $v < 0$ the polarization $\phi_x = \phi_y = 0$, $\phi_z^2 = -r/2(u+v)$ has an Ising configuration, whereas when $v > 0$ the polarization $\phi_x^2 = \phi_y^2 = \phi_z^2 = -r/2(3u+v)$ exhibits diagonal order. The term proportional to v controls the polarization direction in the ferroelectric phase whereas the u term is rotationally invariant. We note that while sweeping v through $v=0$ with $u > 0$ the first-order rotation of polarization direction is accompanied with a continuous change in the magnitude of the polarization. This is driven by a similar mechanism to the Blume-Emery-Griffiths model involving two bosonic fields.²⁰ Within the mean-field approximation the condition for stability of the polarization

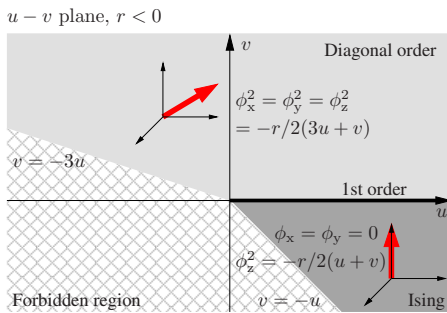


FIG. 1. (Color online) The phase diagram in the u - v plane at zero temperature in the mean-field approximation. The cross-hatched forbidden region denotes where the polarizability would diverge without higher-order corrections. The solid thick line highlights a first-order phase boundary between the light gray region that denotes diagonal order and the dark gray which labels the Ising phase. In each regime the inset axes illustrate the polarization solution highlighted by the bold (red) vector.

is that the net coefficient of the quartic term is positive which translates to $u+v > 0$ when $v < 0$ and $u+v/3 > 0$ if $v > 0$. If these conditions are not fulfilled then higher-order terms must be included and rather than undergo a second-order transition at $r=0$, the system might have a first-order ferroelectric transition at mean-field level. We can neglect the higher order terms such as $\lambda\phi^6$ provided that the model remains stable, which requires that $\lambda\phi^2 \ll u+v$. Here we wish to investigate whether near criticality the fluctuating modes can conspire to drive an otherwise second-order transition to become first order. In order to access this behavior we now go beyond mean field and consider the consequences of quantum fluctuations on the system.

III. FIELD-INTEGRAL FORMULATION

To account for fluctuation corrections to the system Rousev and Millis⁸ employed the renormalization group, which is tailored to study the well-established second-order ferroelectric transition. However, motivated by recent experiments^{1,3} we wish to explore the possibility of a first-order metaelectric transition. Therefore, rather than considering just the corrections due to slow fluctuations that are encompassed by renormalization group, we need to consider fluctuations ψ over all length scales in the polarization $\phi + \psi$ around the saddle-point solution ϕ . When $u \ll r^2$ we can neglect fluctuations in ψ beyond second order which reduces the action to

$$S = \tilde{\beta} \left[\left(r + \frac{g}{3} \right) \phi^2 + u\phi^4 + v \sum_{\alpha} \phi_{\alpha}^4 \right] + \tilde{\beta} \sum_{\tilde{\omega}, \mathbf{q}} \psi^{\dagger}(\tilde{\omega}, \mathbf{q}) G^{-1} \psi(-\tilde{\omega}, -\mathbf{q}), \quad (2)$$

where $G_{\alpha,\beta}^{-1} = G_{\alpha}^{-1} \delta_{\alpha,\beta} + U_{\alpha,\beta}$, the diagonal inverse Green's function takes the form $G_{\alpha}^{-1} = \tilde{\omega}^2 + q^2 + r + f q_{\alpha}^2 + (g - h q^2) q_{\alpha}^2 / q^2 + (4u + 6v) \phi_{\alpha}^2 + 2u\phi^2$, and the off-diagonal terms are $U_{\alpha,\beta} = (g - h q^2) q_{\alpha} q_{\beta} / q^2 + 4u\phi_{\alpha} \phi_{\beta}$. We now integrate over quantum fluctuations to yield the free energy

$$F = \left(r + \frac{g}{3} \right) \phi^2 + u\phi^4 + v \sum_{\alpha} \phi_{\alpha}^4 + \frac{1}{2\tilde{\beta}} \text{Tr} \ln G^{-1}, \quad (3)$$

where $\tilde{\beta} = 1/\tilde{T}$ is the dimensionless inverse temperature. If $\phi=0$ and $r \gg g - h\pi^2$ or if $\phi \neq 0$ and $r \ll \pi^2$ then $UG \ll 1$. In this regime we can expand the inverse Green's function in its off-diagonal terms U using $\text{Tr} \ln G^{-1} = \text{Tr} \ln G^{-1} + \text{Tr} \ln(1 + GU)$ which enables us to describe the renormalization of fluctuations by off-diagonal coupling. This yields

$$F = \left(r + \frac{g}{3} \right) \phi^2 + u\phi^4 + v \sum_{\alpha} \phi_{\alpha}^4 + \frac{1}{\tilde{\beta}} \sum_{\alpha} \left(\text{Tr} \ln \sinh \left[\frac{\tilde{\beta} \xi_{\mathbf{q}}^{\alpha}}{2} \right] - \ln \left[\frac{\tilde{\beta} \xi_{\mathbf{q}}^{\alpha}}{2} \right] \right) - \frac{1}{4\tilde{\beta}} \text{Tr}(UGUG), \quad (4)$$

where $\xi_{\mathbf{q}}^{\alpha} = [q^2 + r + f q_{\alpha}^2 + (4u + 6v) \phi_{\alpha}^2 + 2u\phi^2]^{1/2}$. To remove the fluctuations of the static uniform component of ψ , which

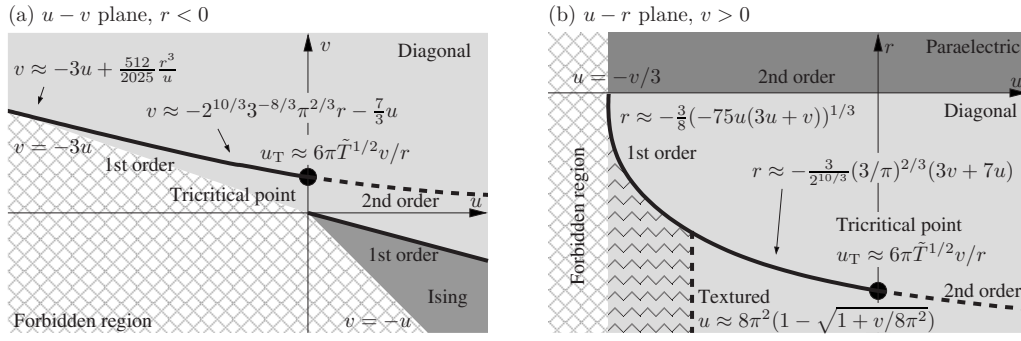


FIG. 2. The phase diagram at $\tilde{T}=0$ in the (a) $u-v$ plane with $r<0$ and (b) $u-r$ plane with $v>0$, both at zero temperature. The cross-hatched forbidden region denotes where the polarizability would diverge without higher-order corrections, the light gray denotes diagonal ordered polarization, and the dark gray the (a) Ising phase and (b) paraelectric phase. Solid thick lines denote first-order phase boundaries, dashed lines second-order transitions, and the circle the tricritical point.

are included in ϕ , we must introduce the second logarithm. This has the effect of regularizing the divergence which would otherwise develop from the first logarithm. This expression, except for the final fluctuation correction term, agrees with that of Ref. 8, and is analogous to the coupling of transverse ferromagnetic fluctuations that led the emergence of first-order behavior.¹⁰ The condition for stability is the same as for the mean-field case.

The momentum integrals are in general evaluated numerically. However, to further investigate the diagonal ordered phase we make the approximation that the cuboid Brillouin-zone boundary ($-\pi < q_\alpha < \pi$) that bounds the momentum space integral can be replaced with a spherical boundary that encloses the same total phase space so has radius $q_D = \sqrt[3]{6\pi^2}$. In the low-temperature limit with the polarization aligned in the (1,1,1) direction, the resulting integrals can then be evaluated analytically to yield

$$\begin{aligned}
 F = & \left(r + \frac{g}{3} \right) \phi^2 + u \phi^4 + v \sum_{\alpha} \phi_{\alpha}^4 \\
 & + \frac{3}{32\pi^2} \left[\pi \sqrt{\xi + \pi^2} (\xi + 2\pi^2) - \xi^2 \ln \left(\frac{\pi}{\sqrt{\xi}} + \sqrt{1 + \frac{\pi^2}{\xi}} \right) \right] \\
 & + \frac{u^2 \phi^2}{6\pi^2} \left[\frac{2\pi}{\sqrt{\xi + \pi^2}} - 2 \ln \left(\frac{\pi}{\sqrt{\xi}} + \sqrt{1 + \frac{\pi^2}{\xi}} \right) \right] \quad (5)
 \end{aligned}$$

with $\xi \equiv r + 2(u+v)\phi^2 + 4u\phi^2/3$ and were found to be in good agreement with the corresponding numerical result.

A. Phase behavior and heat capacity

The phase behavior of the system is shown in Fig. 2. The forbidden region indicates where the action polarizability and free energy would diverge without considering higher-order corrections to the original action. When considered within the framework of mean-field phenomenology, here the system could undergo a first-order paraelectric-ferroelectric transition. However, the corrections due to quantum fluctuations renormalize the action, causing a metaelectric boundary to peel away from the first-order transition associated with the forbidden region. This metaelectric transition is consistent with recent experimental evidence for a first-order phase

transition¹ in ¹⁸O-exchanged SrTiO₃. In both of the planes considered, the line of first-order metaelectric transitions covers an extensive region of the phase diagram, terminating in a tricritical point at $u=0$. The first-order transition at small u is destroyed at nonzero temperature with the tricritical point moving up the line of transitions to $u \approx 6\pi\tilde{T}^{1/2}v/r$. This critical behavior does not depend on the long-range dipole interactions since the lowest-order term in g and h averages to zero on integrating over momenta.⁸ The $\phi \rightarrow -\phi$ symmetry could be destroyed by applying a uniaxial electric field misaligned to the lattice.

A further ramification of the quantum-fluctuation corrections is that the rotation of the polarization from Ising to diagonal order no longer occurs where v turns negative. Though, as for the mean-field case, the magnitude of the polarization is conserved, fluctuations renormalize the quartic terms and shift the phase boundary in Fig. 2(b). This behavior can also be recovered by a renormalization-group analysis.⁸ One experimental probe of the metaelectric transition is the changing behavior of the heat capacity $C = -T\partial^2 F/\partial T^2$. Before the metaelectric transition (small negative r) the relevant optic mode is “soft” and so the heat capacity follows the familiar Debye form $C \sim T^3$ whereas after the metaelectric transition (large negative r), the relevant optic modes are “stiff” and so the heat capacity has an exponential dependence on temperature. At high temperature, in both cases the heat capacity has the expected classical behavior $C = 3k_B$.

Having confirmed the existence of a possible metaelectric behavior, we now turn to consider the stability of the phase in the vicinity of the transition. Recent studies of itinerant ferromagnetism have suggested that such first-order behavior can be preempted by the development of textured magnetic order analogous to that seen in the FFLO phase of superconductors.¹⁵ This leaves open the question as to whether a textured phase can develop in the vicinity of the metaelectric transition. Our strategy to explore this possibility is to assume that the inhomogeneous phase is formed continuously, which allows us to develop a Landau expansion in the polarization Φ and texture wave vector \mathbf{Q} . The onset of an inhomogeneous phase is signaled by the coefficient of the $\Phi^2 Q^2$ term turning negative. In our analysis we search primarily in the vicinity of the metaelectric transition

at $\xi=0$ and consider a trial state with uniform polarization ϕ that is for simplicity superimposed by an inhomogeneous component $\Phi \cos(\mathbf{Q} \cdot \mathbf{r})(1, 1, 1)$. We then expand the free energy to quartic order in \mathbf{Q} and discover that the presence of a textured phase makes a contribution to the total energy of $Q^2\Phi^2[1-u^2\Phi^2/6\pi^2\xi+7u^2\Phi^2Q^2/60\pi^2\xi^2]$. Short of the first-order transition where $\xi < 0$, the coefficient of Q^2 is positive so the phase is not modulated. After the first-order transition ξ turns positive driving the coefficient of Q^2 negative, revealing a finite Q instability in the region highlighted in Fig. 2(b). The modulation carries polarization $\Phi=r/2(u+3/v)$. Though the analysis is restricted to the consideration of a potential continuous transition into the textured phase and a simple form for the texture, it is sufficient to validate its existence. Refinements to include a putative first-order transition or further textured phases would only enlarge the re-

gion of the phase diagram over which inhomogeneities could be observed. Leaving aside potential textured phases we now turn to consider the behavior of the susceptibility across the phase diagram.

B. Inverse susceptibility

The inverse susceptibility provides an experimental window^{3,16,17} onto the quantum-critical properties of ferroelectrics. Deep in the paraelectric regime where $R \equiv r+g/3 \gg q_D$, the contribution to the inverse susceptibility is $\chi^{-1} = \partial^2 F / \partial \phi^2|_{\phi_{\text{eqm}}} = R + \frac{5u+3v}{\pi^2} + \frac{R}{6}(\gamma - \tan^{-1} \gamma) \coth(\frac{\sqrt{R}}{2\tilde{T}})$, which is consistent with Barrett's formula²¹ for a gapped system. In the quantum-critical regime we see three characteristic types of behaviors for the inverse susceptibility

$$\chi^{-1} \approx R + \frac{5u+3v}{\pi^2} \begin{cases} \frac{R}{4}(\gamma\sqrt{1+\gamma^2} - \sinh^{-1}\gamma) + \frac{\pi^2\tilde{T}^2}{18} & \tilde{T} \ll \frac{q_D}{2} \\ \frac{\sqrt{R}}{3}(\gamma - \tan^{-1} \gamma)\tilde{T} & \tilde{T} \gg \frac{q_D}{2} \end{cases} \\ + \begin{cases} 0 & \tilde{T} \ll \frac{\sqrt{g}}{2} \\ \frac{5u+3v}{20} \left(\frac{g}{6} - \sqrt{gh} \right) \tilde{T} & \tilde{T} \gg \frac{\sqrt{g}}{2} \end{cases} \\ + \frac{h^2}{15\pi} \begin{cases} (5u+3v) \left(\frac{3q_D^2}{16} + \frac{\pi^2\tilde{T}^2}{2} \right) & \tilde{T} \ll \frac{q_D}{2} \\ 2q_D\tilde{T} & \tilde{T} \gg \frac{q_D}{2} \end{cases},$$

where the first term is from the diagonal contributions to Eq. (4) and the latter two terms are the off-diagonal contribution, and $\gamma=q_D/R$. At low-temperature the effects of long-range dipole interactions prevail as the off-diagonal fluctuating contribution renormalizes the on-diagonal terms with the linear temperature dependence of the term proportional to g giving a positive slope to the inverse susceptibility whereas the \sqrt{gh} term could provide a negative slope. At higher temperatures the T^2 contribution from the mean-field term dominates, which is also characteristic of quantum-critical behavior and is in good agreement with recent experimental results.³ We note that the T^2 behavior is recovered by other models, including a diagrammatic resummation,^{5,6} the quantum-spherical model,²² renormalization-group studies,^{23,24} a self-consistent phonon model,³ and an analogy to the temporal Casimir effect.⁹ The behavior has also been observed experimentally.^{3,16,17} In both SrTiO₃ and KTaO₃ the initial linear negative slope and the quadratic $\chi^{-1} \sim T^2$ term conspire to cause a characteristic minimum in the inverse susceptibility. Using estimates for the parameters in

Table I, the minimum occurs at $T \sim 1$ K in both SrTiO₃ and KTaO₃ which is in good agreement with the experimental values of $T=1.6$ K and $T=3.0$ K, respectively.³ Finally, at high temperatures a classical term $\chi^{-1} \sim T$ from the longitudinal fluctuating term dominates from ~ 100 K which is again in good agreement with the experimental observations.³

IV. DISCUSSION

In this paper we have found that the polar fluctuating phonons can drive a displacive ferroelectric through a first-order metaelectric transition. Long-range dipolar interactions did not affect this critical-phase behavior.⁸ However, long-range dipole interactions introduced into the action through the term $(g-hq^2)\phi^2$ were pivotal in creating the correction to the inverse susceptibility $\chi^{-1} \sim -T$ that could explain the characteristic inverse susceptibility minimum³ as well as provide important corrections to the self-consistent phonon treatment.³

However, another mechanism, coupling of the soft optic modes to acoustic phonons could be significant. It has already been understood^{3,6,9} that a coupling with the acoustic phonons φ of the form $-\eta(\nabla\varphi)\phi^2$ leads to a correction in χ^{-1} of $-T^4$ that could explain the characteristic minimum in the inverse susceptibility and also has the capability of driving a first-order transition.^{3,9} This work and the results presented here motivate further experimental investigations into the inverse susceptibility and putative metaelectric transition that could shed light on the origin of the phase structure. Though the coupling to acoustic phonons complicated the solid-state system, ultracold atoms in an optical lattice with long-range dipole interactions²⁵ present a clean system that could provide powerful tools to help unravel the properties of the generic Hamiltonian.

One important simplification was to model the ferroelectric with undamped dynamics. Damping would primarily arise due to free electrons, which can be introduced control-

ably through doping. Analogous to “avoided criticality” at a magnetic critical point which leads to non-Fermi-liquid behavior and superconductivity, ferroelectrics might also be expected to adopt novel behavior; for example, doped SrTiO₃,²⁶ whereas undoped SnTe (Ref. 27) and GeTe (Ref. 28) become superconducting at low temperatures. This area presents a promising avenue of research. Further open questions are to determine whether with just a change in parameters^{8,29} the same formalism be applied to order-disorder ferroelectrics and to consider the consequences of the coupling of fluctuating polarization and magnetization that could arise in EuTiO₃.³⁰

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