Quantum Annealed Criticality

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Experimentally there exist many materials with first-order phase transitions at finite temperature that display quantum criticality. Classically, a strain-energy density coupling is known to drive first-order transitions in compressible systems, and here we generalize this Larkin-Pikin[1] mechanism to the quantum case. We show that if the $T = 0$ system lies above its upper critical dimension, the line of first-order transitions can end in a quantum annealed critical point where zero-point fluctuations restore the underlying criticality of the order parameter.

The interplay of first-order phase transitions with quantum fluctuations is an active area [2–9] in the study of exotic quantum states near zero-temperature phase transitions [10–16]. In many metallic quantum ferromagnets, coupling of the magnetization to low energy particle-hole excitations transforms a high temperature continuous phase transition into a low temperature discontinuous one, and the resulting classical tricritical points have been observed in many systems [2–9]. Experimentally there also exist insulating materials that have classical first-order transitions that display quantum criticality [17–21], and here we provide a theoretical basis for this observed behavior.

At a first-order transition the quartic mode-mode coupling of the effective action becomes negative. One mechanism for this phenomenon, studied by Larkin and Pikin [1] (LP), involves the interaction of strain with a fluctuating critical order parameter. LP found that a diverging specific heat in the clamped system of fixed dimensions leads to a first-order transition in the unclamped system at constant pressure. Specifically, the Larkin-Pikin criterion [1, 22] for a first order phase transition is

$$\kappa \; < \; \frac{\Delta C_V}{T_c}\left(\frac{dT_c}{d\ln V}\right)^2 \qquad (1)$$

where V is the volume, ΔC_V is the singular part of the specific heat capacity in the clamped system, T_c is the transition temperature and $\frac{dT_c}{d\ln V}$ is its strain derivative. The effective bulk modulus κ is defined as $\kappa^{-1} = K^{-1} - (K + \frac{4}{3}\mu)^{-1}$ where K and μ are the bare bulk and the shear moduli in the absence of coupling to the order parameter fields; more physically $\kappa \sim K\frac{c_L^2}{c_T^2}$ where c_L and c_T are the longitudinal and the transverse sound velocities [23]. We note that shear strain plays a crucial role in this approach that requires $\mu > 0$. Short-range fluctuations in the atomic displacements renormalize the quartic coupling of the critical modes, but it is the coupling of the uniform ($q = 0$) strain to the energy density, the modulus squared of the critical order parameter, that results in a *macroscopic* instability of the critical point leading to a discontinuous transition.

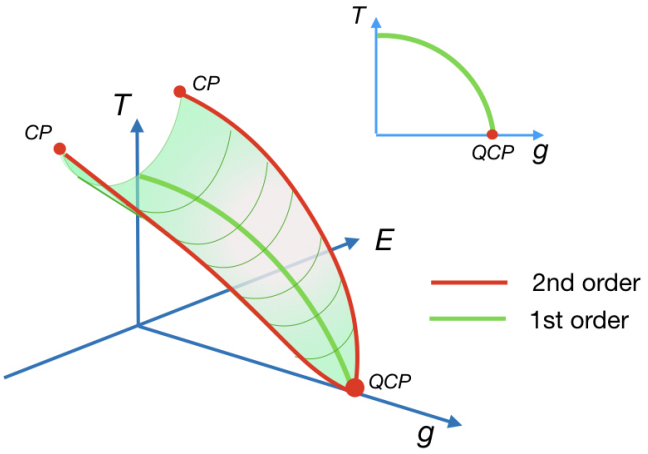

FIG. 1. Proposed Temperature (T)-Field (E)-Pressure (g) Phase Diagram with a sheet of first-order transitions bounded by second-order phase lines linking the three critical points, two classical and one quantum; here g (e.g. Pressure) tunes the quantum fluctuations and E (e.g. Electric Field) is the field conjugate to the order parameter (e.g. Polarization). Inset: Temperature-Pressure "slice" indicating a line of classical phase transitions ending in a "quantum annealed critical point" with the standard temperature fan where the underlying order parameter criticality is restored by zero-point fluctuations.

Here we rewrite the Larkin-Pikin criterion in terms of correlation functions so that it can be generalized to the quantum case. We show that if the $T = 0$ quantum system lies above its upper critical dimension, the corrections to the renormalized bulk modulus are non-universal; the line of classical first-order transitions can end in a "quantum annealed critical point" where zero-point fluctuations restore the underlying criticality of the order parameter. We end with a discussion of the temperature-field-pressure phase diagram and specific measurements to probe it (cf. Fig. 1).

Low-temperature measurements on ferroelectric insulators provide a key motivation for our study [17–21]. At finite temperatures and ambient pressure these ma-

terials often display first-order transitions due to strong electromechanical coupling [24]; yet in many cases [17–21] their dielectric susceptibilities suggest the presence of pressure-induced quantum criticality associated with zero-temperature continuous transitions [17–21]. It is thus natural to explore whether a quantum generalization of the Larkin-Pikin approach [1], involving the coupling of critical order parameter fluctuations to long wavelength elastic degrees of freedom, can be developed to describe this phenomenon.

In the simplest case of a scalar order parameter ψ and isotropic elasticity, the Larkin-Pikin (LP) mechanism [25] refers to a system where the order parameter $\psi(\vec{x})$ is coupled to the volumetric strain with interaction energy

$$H_I = \lambda \int d^3x \; e_{ll}(\vec{x}) \; \psi^2(\vec{x}) \tag{2}$$

where $e_{ab}(\vec{x}) = \frac{1}{2}\left(\frac{\partial u_a}{\partial x_b} + \frac{\partial u_b}{\partial x_a}\right)$ is the strain tensor, $u_a(\vec{x})$ is the atomic displacement, $e_{ll}(x) = \mathrm{Tr}[e(\vec{x})]$ is the volumetric strain and λ is a coupling constant associated with the strain-dependence of T_c, $\lambda = \left(\frac{dT_c}{d\ln V}\right)$. Though the elastic degrees of freedom are assumed to be Gaussian, and thus can be formally integrated out exactly, this must be done with some care. This is because the strain field separates into a uniform ($\vec{q} = 0$) term defined by boundary conditions and a finite-momentum ($\vec{q} \neq 0$) contribution determined by fluctuating atomic displacements

$$e_{ab}(\vec{x}) = e_{ab} + \frac{1}{V}\sum_{\vec{q}\neq 0} \frac{i}{2}[q_a u_b(\vec{q}) + q_b u_a(\vec{q})]e^{i\vec{q}\cdot\vec{x}}, \tag{3}$$

where $\{a, b\} \in [1, 3]$ and $u_a(q)$ is the Fourier transform of $u_a(x)$. Here we employ periodic boundary conditions to a finite size system with volume $V = L^3$ and discrete momenta $\vec{q} = \frac{2\pi}{L}(l, m, n)$, where l, m, n are integers.

The uniform strain vanishes when the crystal is externally clamped. The main effect of integrating out the finite wavevector fluctuations in the strain is to induce a finite correction to the short-range interactions of the critical fluctuations that can be absorbed into the quartic ψ^4 terms in the action. By contrast, fluctuations in the uniform component of the strain induce an infinite-range attractive interaction between the critical modes (see Supplementary Materials), and it is this component of the interactions that is responsible for driving first order behavior. The problem is then reduced to the interaction of critical order parameter modes, mediated by the fluctuations of a uniform strain field ϕ with bulk modulus κ (for details see Supplementary Materials). Conceptually, the Larkin-Pikin approach amounts to a study of critical phenomena in a clamped system, followed by a stability analysis of the critical point once the clamping is removed.

Recently it was proposed to adapt the Larkin-Pikin approach to pressure($\mathcal{P}$)-tuned quantum magnets where it is often found that $\frac{dT_c}{d\mathcal{P}} \to \infty$ as $T_c \to 0$; the authors argued that the associated quantum phase transitions should then be first-order [26–28]. However such a diverging coupling of the critical order parameter fluctuations and the lattice should lead to structural instabilities near the quantum phase transition that have not been observed [9, 29]. Furthermore dynamics must be included when treating thermodynamic quantities at zero temperature [30, 31].

We recast the Larkin-Pikin criterion in the language of correlation functions, generalizing the LP approach to the quantum case summing over all possible space-time configurations. The strain field again separates into two contributions as in equation (3), one associated with static uniform boundary conditions and the other determined by short wavelength displacements fluctuating at all frequencies

$$e_{ab}(\vec{x},\tau) = e_{ab} + \frac{1}{\beta V}\sum_{i\nu_n}\sum_{\vec{q}\neq 0} \frac{i}{2}[q_a u_b(q) + q_b u_a(q)]e^{i(\vec{q}\cdot\vec{x} - \nu_n\tau)} \tag{4}$$

where $q_\alpha \equiv (\vec{q}, i\nu_n)$ with $\alpha \in [1, 4]$, $u_b(q) \equiv u_b(\vec{q}, i\nu_n)$ and $\nu_n = 2\pi n T$ is a Matsubara frequency ($k_B = 1$). A detailed analysis indicates that when these space-time elastic degrees of freedom are integrated out, they lead to the coupling of the quantum critical order parameter modes to a *classical* strain field ϕ, uniform in both space and time, with the same effective bulk modulus κ as in the finite-temperature case (see Supplementary Material). The resulting effective action takes the form

$$S_{eff}[\psi, \phi] = \int_0^\beta d\tau \int d^3x \left[\mathcal{L}[\psi] + \lambda\phi \; \psi^2(\vec{x},\tau) + \frac{1}{2}\kappa\phi^2\right], \tag{5}$$

where $(\vec{x}, \tau)$ are the Euclidean space-time co-ordinates and $\mathcal{L}[\psi]$ is the Lagrangian of the order parameter $\psi(\vec{x}, \tau)$ that undergoes a continuous transition in the clamped system; in the simplest case $\mathcal{L}[\psi]$ is a ψ^4 field theory

$$\mathcal{L}[\psi] = \frac{1}{2}(\partial_\mu\psi)^2 + \frac{a}{2}\psi^2 + \frac{b}{4!}\psi^4. \tag{6}$$

The partition function of the unclamped system is then

$$Z[\phi] = e^{-\beta F[\phi]} = \int \mathcal{D}[\psi] e^{-S_{eff}[\psi,\phi]}, \tag{7}$$

where the trace is over the internal variable ψ, and $Z[\phi]$ to be evaluated at the stationary point $F'[\phi] = 0$. The renormalized bulk modulus, $\tilde{\kappa} = \kappa - \Delta\kappa$, is

$$\tilde{\kappa} = \frac{1}{V}\frac{\partial^2 F}{\partial\phi^2} = \kappa - \lambda^2 \int d^3x d\tau \langle \delta\psi^2(\vec{x},\tau)\delta\psi^2(0)\rangle, \tag{8}$$

where $\delta\psi^2(\vec{x},\tau) = \psi^2(\vec{x},\tau) - \langle\psi^2\rangle$. In the classical problem there is no time-dependence, and $\int_0^\beta d\tau \to \beta \equiv 1/T$

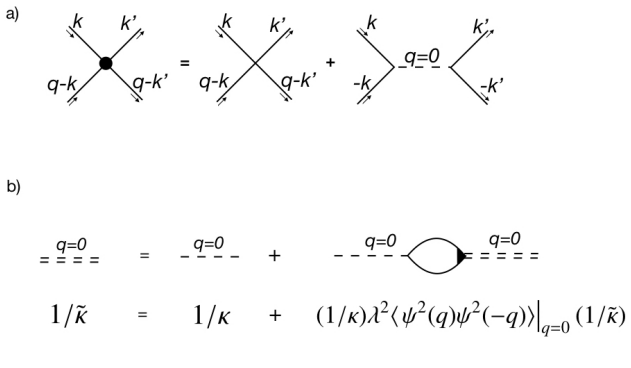

FIG. 2. Diagrammatic approach to the generalized Larkin-Pikin criterion a) Bare interaction is a sum of a local and a nonlocal contribution mediated by fluctuations in the strain; b) Feynman diagram showing renormalization of the strain propagator by coupling to energy fluctuations.

so at the transition

$$\tilde{\kappa} = \frac{1}{V}\frac{\partial^2 F}{\partial \phi^2} = \kappa - \frac{\lambda^2}{T_c}\int d^3x \langle \delta\psi^2(\vec{x})\delta\psi^2(0)\rangle = \kappa - \Delta\kappa. \tag{9}$$

$\Delta\kappa$ in (9) is proportional to energy fluctuations, and can be re-expressed as $\frac{\lambda^2}{T_c}\Delta C_V$; we thus recover the LP criterion (1) ($\kappa < \Delta\kappa$ or $\tilde{\kappa} < 0$) for a first-order transition.

The renormalized bulk modulus $\tilde{\kappa}$ can also be obtained diagrammatically (cf. Figure 2). In the low-energy effective action, the quartic term now has a contribution from the coupling of the order parameter fluctuations to the effective uniform strain. We then can use a Dyson equation for the strain propagator to determine $\tilde{\kappa}$. More specifically we can write

$$\left(\frac{1}{\tilde{\kappa}}\right) = \left(\frac{1}{\kappa}\right) + \left(\frac{1}{\kappa}\right)\lambda^2\langle\psi^2(q)\psi^2(-q)\rangle\big|_{q=0}\left(\frac{1}{\tilde{\kappa}}\right) \tag{10}$$

that results in

$$\tilde{\kappa} = \kappa - \Delta\kappa = \kappa - \lambda^2\chi_{\psi^2} \tag{11}$$

where $\chi_{\psi^2} = \chi_{\psi^2}(\vec{q}, i\nu_n)\big|_{\vec{q},i\nu_n=0}$ is the static susceptibility for ψ^2, where

$$\chi_{\psi^2}(\vec{q}, i\nu_n) = \int_0^\beta d\tau \int d^dx \langle \delta\psi^2(\vec{x},\tau)\delta\psi^2(0)\rangle e^{i\nu_n\tau - i\vec{q}\cdot\vec{x}}, \tag{12}$$

is the Fourier transform of the fluctuations in ψ^2 and $d = 3$. The sign of $\tilde{\kappa}$ in (11) is determined by the infrared behavior of $\Delta\kappa$; if it diverges, as it does classically (for a scalar order parameter and isotropic elasticity), then this correction is universal and the transition is first order.

Another possibility is revealed in the zero-temperature long-wavelength Gaussian approximation of (11). If we make the Gaussian approximation $\langle\delta\psi^2(x)\delta\psi^2(0)\rangle \approx (\langle\delta\psi(x)\delta\psi(0)\rangle)^2$, then

$$\lim_{T\to 0}\Delta\kappa \propto \int dq\ d\nu\ q^{d-1}\ [\chi_\psi(\vec{q}, i\nu)]^2 \tag{13}$$

where $\chi_\psi(\vec{q}, i\nu)$, the order parameter susceptibility, is the Fourier transform of the correlator $\langle\psi(x)\psi(0)\rangle$. Since dimensionally $[\chi] = \left[\frac{1}{q^2}\right]$ and $[\nu] = [q^z]$, we find that in the approach to the quantum phase transition

$$\lim_{T\to 0}[\Delta\kappa] = \frac{[q^{d+z}]}{[q^4]} \tag{14}$$

so that the quantum corrections to κ are non-singular for $d + z > 4$. The presence of quantum zero-point fluctuations increases the effective dimensionality of the phase space for order parameter fluctuations. If the effective dimensionality of the quantum system lies above its upper critical dimensionality, this will have the effect of liberating the quantum critical point from the inevitable infrared slavery experienced by its finite-temperature classical counterpart. In particular the correction to the renormalized bulk modulus is then non-universal, allowing for quantum annealed criticality where zero-point fluctuations toughen the system against the macroscopic instability present classically, restoring its underlying continuous phase transition.

We have therefore identified a theoretical scenario where there is a quantum continuous transition even though all transitions at finite temperature are first-order. Application of a field conjugate and parallel/antiparallel to the order parameter in such a system leads to a line of first-order transitions ending in two classical critical points. Therefore by continuity there is a surface of first-order phase transitions in the phase diagram (cf. Figure 1) connecting the three critical points, one quantum and two classical, bounded by second-order phase lines. This phase diagram then presents an alternative scenario of the interplay of discontinuous transitions and fluctuations to that studied in metallic magnets where applied field is needed to observe quantum criticality in addition to the tuning parameter [9].

The specific heat exponent α plays a key role in the universality of the classical Larkin-Pikin criterion (1) since the coupling of the order parameter to the lattice is a strain-energy density. For the scalar ($n = 1$) case considered here, $\alpha > 0$, so that $\Delta\kappa$ is singular and the finite-temperature transition is always first-order; for $d+z > 4$, there is a quantum annealed criticality but no quantum tricritical point since the quartic mode-mode term in the effective action jumps from negative to positive due to the change of effective dimension.

Because of its underlying non-universal nature, the possibility of pressure-tuned quantum annealed criticality must be determined in specific setttings. Many ferroelectrics have scalar order parameters with dynamical exponent $z = 1$, so such three-dimensional materials are

in their marginal dimension; logarithmic corrections to the bulk modulus are certainly present but they are not expected to be singular. Indeed such contributions to the dielectric susceptibility, χ, in the approach to ferroelectric quantum critical points have not been observed to date [20]; furthermore here the temperature-dependence of χ is described well by a self-consistent Gaussian approach appropriate above its upper critical dimension [20, 21]. Therefore there may be a very weak first-order quantum phase transition [32] but experimentally it appears to be indistinguishable from a continuous one. We note that near quantum criticality the main effect of long-range dipolar interactions, not included in this treatment, is to produce a gap in the logitudinal fluctuations, but the transverse fluctuations remain critical [33–35]; the excellent agreement between theory and experiment at ferroelectric quantum criticality confirms that this is the case [20, 21].

Dielectric loss and hysteresis measurements can be used to probe the line of classical first-order transitions, and to determine the nature of the quantum phase transition. The Gruneisen ratio (Γ), the ratio of the thermal expansion and the specific heat, is known to change signs across the quantum phase transition [36, 37]; furthermore it is predicted to diverge at a 3D ferroelectric quantum critical point as $\Gamma \propto \frac{1}{T^2}$ so this would be a good indicator of underlying quantum criticality [21]. Both the bulk modulus and the longitudinal sound velocity should display jumps near quantum annealed criticality, though specifics are material-dependent since the fluctutation contributions to both are non-universal.

For systems with multi-component order parameters ($n \geq 2$), the specific heat exponent α is negative so the correction to the renormalized bulk modulus will be nonuniversal even at finite temperatures [22, 38, 39]. In this case, there can be a classical tricritical point at finite pressures with a second-order transition that continues to zero temperature; this situation should be robust to everpresent disorder following the Harris criterion [40]. By contrast everpresent elastic anisotropy is known to destabilize criticality in the classical isotropic elastic scalar ($n = 1$) lattice and to drive it first-order into an inhomogeneous state [22, 38, 39]; here quantum annealed criticality may still be possible due to the increase of effective dimensionality. The coupling of domain dynamics to anistropic strain has been studied classically for ferroelectrics [41], and implications for the quantum case are a topic for future work.

In summary, we have developed a theoretical framework to describe compressible insulating systems that have classical first-order transitions and display pressure-induced quantum criticality. We have generalized the Larkin-Pikin criterion [1] in the language of correlation and response functions; from this standpoint it is clear that the correction to the renormalized bulk modulus, singular at finite temperature, is non-universal at $T = 0$ for $d + z > 4$ so then the quantum transition may be continuous. Our analyis has been performed for the case of a scalar order parameter and isotropic elasticity where the phase transition is first-order for all finite temperature; in this extreme instance we argue that it is still possible to have quantum annealed criticality. Naturally the presence of a finite-pressure classical tricritical point ensures a continuous quantum phase transition. The key point is that a compressible material can host a quantum critical phase even if it displays a first-order transition at ambient pressure. More generally the order of the classical phase transition can be different from its quantum counterpart.

We note in ending that there are experiments on metallic systems [42–44] that also suggest quantum annealed criticality, so a quantum generalization of the electronic case [45] with possible links to previous work on metallic magnets should be pursued [9]; implications for doped paraelectric materials and polar metals [21] will also be explored. Extension of this work to quantum transitions between two distinct ordered states separated by first-order classical transitions may be relevant to the iron-based superconductors [46] and to the enigmatic heavy fermion material URu_2Si_2 where quantum critical endpoints have been suggested [47]. Finally the possibility of quantum annealed criticality in compressible materials, magnetic and ferroelectric, provides new settings for the exploration of exotic quantum phases where a broad temperature range can be probed with easily accessible pressures due to the lattice-sensitivity of these systems.

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