Classical and quantum local criticality due to electron-vibration interaction

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We study the local classical and quantum critical properties of electron-vibration interaction, represented by the Yu-Anderson model. It exhibits an instability, similar to the Wentzel-Bardeen singularity, whose nature resembles to weakly first-order quantum-phase transitions at low temperatures, and crosses over to Gaussian behavior with increasing temperature. We determine the dominant energy scale separating the quantum from classical criticality, study the effect of dissipation, and analyze its impact on correlation functions. Similar phenomenon should be observable in carbon nanotubes around local defects.

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I. INTRODUCTION

Quantum-phase transitions are intensively studied due to the governing fundamental physical properties and also because a number of highly interesting systems display such behavior. These occur at zero temperature at quantum critical points (QCPs) and are dominated by quantum rather than classical fluctuations. Their study enriches our knowledge on classical and quantum critical behavior and can reveal the connections between them in terms of quantum-to-classical mappings.[1] Quantum-phase transitions were found to explain the behavior of Ge-doped $YbRh_2Si_2$ (Ref. 2) and other heavy fermion materials.[3]

The notion "local criticality" stands for quantum-phase transitions in, e.g., quantum impurity models which evolve only in time but are confined in space, such as the quantum-phase transition in the sub-Ohmic and Ohmic spin-boson model,[4,5] the dissipation-induced phase transition in a quantum box,[6] or the local quantum phase transition in the pseudogap Anderson model.[7]

Local vibrational modes due to foreign (adsorbed) molecules or lattice imperfections strongly influence the electronic transport and induce dephasing through inelastic scattering.[8] Molecular electronic devices[9,10] are probed and controlled locally by single-molecule vibrational spectroscopy[11] based on scanning tunneling microscope (STM) and inelastic electron tunneling spectroscopy and the spectrum of molecular vibrations often indicates more complex behavior than can be seen in bulk transport[12] such as negative differential resistance and hysteresis.[13] Conductance measurements on mechanically controllable break junctions reveal the presence of local vibrational degrees of freedom, when noble-metal (Pt) electrodes were connected by a single molecule as Pt or H_2.[14,15] The vibrational mode softens after coupling it to electrons, and the vibrational resonance in the conductance due these modes shifts to lower energies. A critical bosonic mode caused by the interplay of softening and dissipation contributes to transport down to very low temperatures. Thus, the detailed understanding of these modes and their local criticality beyond the mean field are essential,[12] since even away from the critical point, they leave their mark on the responses.[1]

Recently, the observation of strong phonon modes were reported in suspended carbon nanotubes (CNs) by STM.[10] The electronic properties of CN are tunable by chemical doping or by changing the chirality of the tube, hence these systems are ideal candidates to study and control enhanced molecular vibrations,[9,10] developing around lattice imperfections or encapsulated molecules via a local deformation potential. Criticality due to the local electron-vibration interaction is regarded as the descendant of the Wentzel-Bardeen (WB) singularity,[16–18] which arises in a one-dimensional system of electrons, coupled to long-wavelength phonons, which only allows for forward scattering. For a critical value of the electron-phonon coupling, the system becomes unstable and acquires a negative compressibility. The thermodynamic quantities and correlation functions for the electron-phonon system were studied near this singular point,[19,20] indicating the presence of a phase transition. The divergence of the specific heat is accompanied by a collapse of the system induced by the strong electron-phonon interaction. For a critical value of the electron-phonon coupling, the system becomes unstable and acquires a negative compressibility. A recent study[21] suggested that the WB singularity could be reached experimentally in thick carbon nanotubes due to phonons.

Local vibrational modes and their critical properties are interesting for a variety of other reasons. Critical modes are important in spintronics since they unavoidably lead to decoherence even at very low temperatures. Electron-vibration interactions are exceptionally important for molecular solids where highly energetic vibrational states greatly influence the electronic properties. Examples include the superconductivity with $T_c=40$ K in fullerides[22] and the energetic electron-phonon sidebands in the excitonic excitation states of single-wall carbon nanotubes.[23]

This paper addresses the local criticality caused by the electron-vibration interaction. We determine the critical exponents and show that a weakly first-order quantum-phase transition governs the low-temperature physics, similarly to certain Ising models. At high temperatures, it crosses over to classical Gaussian behavior. This crossover influences the electronic properties as well, e.g., in carbon nanotubes, which can be revealed by local spectroscopical measurements.[11]

II. MODEL AND ITS BASIC PROPERTIES

As schematization of local electron-vibration interaction, we start with the Yu-Anderson or single impurity Holstein model,[24]

$$H = \sum_k \varepsilon(k) c_k^+ c_k + g_d Q \Psi^+(\mathbf{0})\Psi(\mathbf{0}) + \frac{P^2}{2m} + \frac{m\omega_0^2}{2} Q^2, \quad (1)$$

which describes d-dimensional electrons interacting with a local bosonic mode at a single impurity site with position Q and momentum P. The model can be mapped onto one-dimensional chiral fermions interacting with a single vibrational mode, and the fermionic field can be bosonized.[8,25] Then, we arrive to an effective model of one dimensional-coupled harmonic oscillators, i.e., the Caldeira-Leggett (CL) model,[5,26]

$$H = v_c \int_{-\infty}^{\infty} dx [\partial_x \Phi(x)]^2 + \frac{g}{\sqrt{\pi}} Q \partial_x \Phi(0) + \frac{P^2}{2m} + \frac{m\omega_0^2}{2} Q^2, \quad (2)$$

v_c is the charge velocity, and g is the phase shift caused by g_d, $\Phi(x)$ stems from the bosonic representation of the fermion field. This also represents the effective model for the large spin-boson model.[27] After integrating out the bosonized electron field $\Phi(x)$, the effective action for the phonon reads as

$$S_{\mathrm{ph}} = \frac{m}{2T} \sum_n \left[\omega_n^2 + \omega_0^2 \left(1 - \frac{\Gamma}{\Gamma_2} \right) + 2|\omega_n|\Gamma \right] |Q_n|^2, \quad (3)$$

where $\omega_n = 2\pi n T$ is the bosonic Matsubara frequency, Q_n's are the Fourier components of $Q(\tau)$. The main difference with respect to CL is the potential renormalization (the $-\omega_0^2 \Gamma/\Gamma_2$ term), which is avoided in CL to study the effect of pure dissipation. In our case, the phonon is expected to soften after coupling it to electrons on physical ground, therefore, such local term is present in the action.[5] As the phonon-mode softens, its eigenfrequencies on the real frequency axis are given by[25]

$$\omega_{p\pm} = -i\Gamma \pm \sqrt{\omega_0^2(1 - \Gamma/\Gamma_2) - \Gamma^2}, \quad (4)$$

where $\Gamma_2 = \pi\omega_0^2/4W \ll \omega_0 \ll W$, W is the bandwidth of the conduction electrons, and $\Gamma = \pi(g\rho)^2/2m$ for small g, and approaches Γ_2 as $g \to \infty$. Here, $\rho = 1/2\pi v_c$ is the chiral electron density of states. The explicit dependence of Γ on g_d cannot be determined by the bosonization approach.[25] The real part of the phonon frequency remains finite (underdamped) for $\Gamma < \Gamma_1 \approx \Gamma_2(1 - \Gamma_2^2/\omega_0^2)$. For $\Gamma_1 < \Gamma < \Gamma_2$, the oscillatory behavior disappears from the phononic response (Re $\omega_{p\pm} = 0$), and two distinct dampings characterize it (overdamped).

III. QUANTUM CRITICALITY

Close to Γ_2, the softening of the phonon frequency occurs as

$$\omega_{p+} = -\frac{i\omega_0^2}{2\Gamma_2} y, \quad (5)$$

where $y > 0$ is the distance from criticality (the effective reduced "temperature"),

$$y = 1 - \frac{\Gamma}{\Gamma_2}, \quad (6)$$

and following Ref. 1, the characteristic energy scale is expected to vanish as $\omega_{p+} \sim y^{\nu}$. This defines $\nu = 1$, and using[28] $\nu = 1/y_t$, the "thermal" scaling exponent is $y_t = 1$. The extra i in Eq. (5) signals the dissipative nature of the transition. Note that the criticality is tuned by Γ and not by the temperature, so y_t belongs to Γ. At the same time, ω_{p-} approaches $-2i\Gamma_2$.

Since the problem is effectively one dimensional, possesses "zero" spatial dimension, and evolves only in time ($z = 1$), this allows us to set $d_{\mathrm{eff}} = d + z = 1$. The dynamical exponent z can be melted in the definition of d, since there are no separate spatial and temporal dimensions.

To proceed with the exploration of the critical properties of our model, we evaluate its free energy. Using Eq. (3) or following Ref. 25, it is obtained as

$$F = F_e + 2\Gamma + \frac{T}{\pi} \int dx \ln \left| 1 - \exp\left(-\frac{x}{T} \right) \right| \frac{\Gamma(x^2 + \omega_0^2 y)}{(x^2 - \omega_0^2 y)^2 + (2\Gamma x)^2}, \quad (7)$$

where F_e is the phonon free contribution of electrons. Focusing on the most singular contribution in y at $T = 0$, we get

$$f_s(T \to 0) = \frac{\omega_0^2}{4\pi\Gamma_2} y \ln\left(\frac{1}{y} \right) \sim y^{2-\alpha}, \quad (8)$$

leading to $2 - \alpha = 1 = d_{\mathrm{eff}}/y_t$ [using $\ln(x) = \lim_{\varepsilon \to 0}(x^{\varepsilon} - 1)/\varepsilon$], from which $y_t = 1$ is deduced in accordance with the exponent we got from the vanishing of the characteristic energy scale in Eq. (5). This suggests the emergence of a weakly first-order quantum-phase transition.[29] Interestingly, many experimentally observed quantum-phase transitions belong to this category.[30] In contrast to true first-order transitions, where $f_s \sim |y|$, we have logarithmic corrections in y, causing the second derivative of f_s (the equivalent of the specific heat) to diverge in a power-law fashion as $1/y$. True first-order transitions are not accompanied by critical fluctuations. However, the weakly first-order nature of the transition here allows for criticality to develop, which leaves its mark on the fluctuations. The mean square of the Q field follows from the effective action as

$$\langle Q^2 \rangle = \frac{T}{m} \sum_n \frac{1}{\omega_n^2 + \omega_0^2 y + 2\Gamma|\omega_n|}, \quad (9)$$

and diverges at $T = 0$ as $\langle Q^2 \rangle \sim |\ln(y)|$. The weak divergence of the fluctuation is a direct consequence of the weakly first-order nature of the instability at $T = 0$. At finite temperatures, this crosses over to T/y type of divergence, as we discuss below.

TABLE I. Summary of the critical exponent in the quantum and classical critical regions, η is only determined through $\eta=d_{\text{eff}}+2-2y_h$.

	d_{eff}	y_t	y_h	α	β	γ	δ	ν	η
Quantum ($T<T^*$)	1	1	1	1	0	1	∞	1	1
Classical ($T>T^*$)	0	2	1	2	$-1/2$	1	-1		

By coupling an external field (V) to the position as VQ, we can study the resulting distortion within linear response. According to scaling,[31]

$$\langle Q\rangle = V^{1/\delta}\Phi_Q\left(\frac{y}{V^{1/\beta\delta}}\right), \tag{10}$$

where $\Phi_Q(x)$ is a scaling function. From Eq. (3), we get

$$\langle Q\rangle = \frac{V}{m\omega_0^2 y} \sim V^0\left(\frac{y}{V}\right)^{-1}, \tag{11}$$

from which we deduce $\beta=1/\delta=0$. The scaling function is determined as $\Phi_Q(x)=1/x$. This turns out to be close to that of the one-dimensional Ising model.[31]

The divergence of the order-parameter susceptibility defines the γ exponent. In the present case, the phonon-field Q is expected to fluctuate close to the critical-coupling Γ_2 and plays the major role in the instability of the system. Its susceptibility is obtained as

$$\chi_Q = \frac{1}{m\omega_0^2}\frac{1}{y} \sim y^{-\gamma}, \tag{12}$$

with $\gamma=1$. This divergence is analogous to the diverging compressibility at the WB singularity.[19,20] It gives for the exponent of the external field[28]

$$y_h = \frac{y_t\gamma + d + z}{2} = 1. \tag{13}$$

Therefore, the universality class of this problem is defined by $d_{\text{eff}}=y_t=y_h=1$, as is summarized in Table. I.

IV. RELATION TO THE ISING MODEL

Similar exponents characterize the one-dimensional classical ferromagnetic Ising model as well. There, for any real h (longitudinal magnetic field) and finite T, there is no sign of criticality. However, at $T=h=0$, the correlation length diverges, indicating the presence of a critical point. Following the ideas of scaling, the exponents were determined[31] as $\beta=1/\delta=0$, $\alpha=\gamma=\nu=\eta=1$, similarly to what we find here, and the spatial dimension of this classical model is $d=1$. Interestingly, this criticality can be brought to finite temperature, if we consider an infinitely strong ferromagnetic chain or a line of defects between two neighboring columns[32,33] in the two-dimensional Ising model on a square lattice, as shown in Fig. 1. The critical behavior on the defect chain differs from the bulk critical behavior, e.g., the critical exponents vary with the defect chain strength. In the case of a chain with infinitely strong ferromagnetic coupling, the defect contribution to the specific heat diverges[34] with $\alpha=1$ and the order-parameter exponent on the defect chain is $\beta=0$,[32,35] and the other local critical exponents also agree with that of a one-dimensional ferromagnetic Ising chain at $T=0$. Note that the transition temperature of the two-dimensional Ising model is not affected by the presence of defect line. In this respect, the effective one-dimensional critical behavior of the defect is embedded in a two-dimensional critical region, which facilitates its observation.

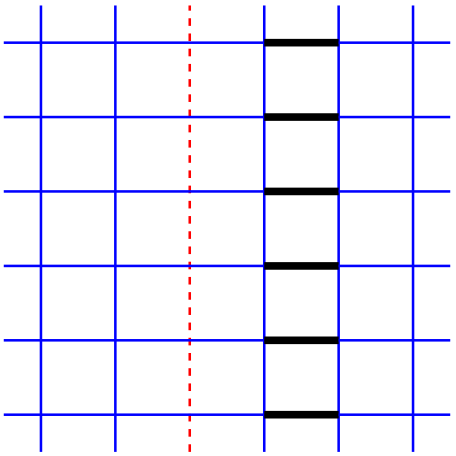

FIG. 1. (Color online) Two types of defect chains (red dashed and thick black lines) are shown for the two-dimensional Ising model, leading to the same local critical behavior.

V. CORRELATION FUNCTIONS

The appearance of quantum criticality is further corroborated by investigating the time evolution of the correlator of the phonon field $\langle Q(\tau)Q\rangle$. The mapping of our d-spatial dimensional quantum system (with $d=0$) onto a $d+1$-dimensional classical one is done in the imaginary time path-integral formalism. There, one introduces an extra imaginary time dimension with size $1/T$, defining the length of the classical counterpart. Thus, as long as the correlation length $\xi<1/T$, the system exhibits the previous $d+1$-dimensional quantum critical behavior. However, for $\xi>1/T$, finite-size effects are important, quantum effect can be neglected, and the system crosses over to d-dimensional classical problem.[36]

At $T=0$, the decay of the correlator in imaginary time is obtained from the action Eq. (3) as

$$G_Q(\tau) = \langle Q(\tau)Q\rangle \sim \int_{-\infty}^{\infty}\frac{\cos(\omega\tau)d\omega}{2\Gamma_2|\omega| + \omega_0^2 y} = \int_0^{\infty}\frac{\Gamma_2^{-1}\cos(x)dx}{|x| + \omega_0^2 y\tau/2\Gamma_2} \sim \begin{cases} (y\tau)^{-2} & \text{for } \tau \gg 2\Gamma_2/\omega_0^2 y \\ -\ln(y\tau) & \text{for } \tau \ll 2\Gamma_2/\omega_0^2 y, \end{cases} \tag{14}$$

and Eq. (14) is valid down to $\tau\sim 1/2\Gamma_2$, where the ω_{p-} frequency starts to play its role. Scaling predicts[28] that

$$G_Q(\tau) = |y|^{2(z-y_h)/y_t}\Phi_\tau\left(\frac{\tau}{y^{-z/y_t}}\right) = \Phi_\tau(\tau y), \tag{15}$$

where $\Phi_\tau(x)$ is a scaling function, and the last relation is obtained for our specific model. From this, we can draw several important conclusions. First, the scaling form predicts a crossover time, separating the long and short τ regions, to scale as $\tau^* \sim 1/y \sim y^{-\nu}$, giving $\nu=1$, in agreement with $\nu=1/y_t$. This is in perfect agreement with the second integral of Eq. (14), where only the $\omega_0^2 y\tau/2\Gamma_2$ combination contains τ and y, suggesting

$$\tau^* = \frac{2\Gamma_2}{\omega_0^2 y}. \tag{16}$$

By approaching the instability ($y \to 0$), $\tau^* \to \infty$. Second, scaling does not predict additional multiplicative powers of y in front of the scaling function, in nice agreement with the analytical result of Eq. (14). Third, the universal scaling function can be determined from Eq. (14): $\Phi_\tau(x)$ decreases as $-\ln(x)$ for $x \ll 1$, and decays algebraically as $1/x^2$ for $x \gg 1$. The former is characteristic to a quantum Brownian particle,[5] which dominates the response when $\tau^* \to \infty$. The latter corresponds to the correlator of a damped harmonic oscillator. As we approach $y \to 0$, the harmonic potential flattens and disappears. Then, our particle does not experience any confinement and performs quantum Brownian motion, which is not bounded, and $\langle Q^2 \rangle$ diverges. Instead of the position Q, the displacement $Q(\tau)-Q(0)$ keeps track of its dynamics.[5]

The electrons also experience criticality similarly to the oscillator. The local charge susceptibility diverges as $1/y$ similarly to the phononic response.[25] The local Green's function of the electrons $G_e(\tau)=\langle \Psi^+(\mathbf{0},\tau)\Psi(\mathbf{0},0)\rangle$ decays as $1/\tau$ for long τ at $T=0$ as in a local Fermi liquid; but for $\tau < \tau^*$, it changes to

$$G_e(\tau) \sim \frac{1}{2\tau}\left[1 - \left(\frac{\tau}{\tau^*}\right)^2 \exp(2\gamma_E)\right], \tag{17}$$

where $\gamma_E \approx 0.577$ is the Euler's constant. Thus, half of the spectral weight is lost at short times due to the scattering off the phonon close to the critical point $y=0$. Consequently, the spectral function [the Fourier transform of $G_e(\tau)$] takes half of its noninteracting value for frequencies larger than $1/\tau^*$. This harmonizes with the behavior of the inelastic scattering rate of the electrons,[8] which reaches its maximal value at $y \to 0$ for high-energy electrons. The above results are valid for $\tau \gg 1/2\Gamma_2$, below which the other mode (ω_{p-}) dominates, similarly to Eq. (14).

By increasing the temperature, it is better to work in real times since τ is restricted in a finite slab. At $T>0$, $G_Q(t)$ decays exponentially with a correlation time given by $\xi_t = 1/2\pi T$. This defines the quantum critical region, when besides temperature, there is no other relevant energy scale in the problem. The diverging ξ_t at $T \to 0$ agrees with the algebraic decay of Eq. (14) at $T=0$. With increasing temperature, we cross over to the classical critical region at

$$T^* = \frac{\omega_0^2}{4\pi\Gamma_2} y. \tag{18}$$

For $T > T^*$, the coherence time becomes $\xi_t = 2\Gamma_2/\omega_0^2 y$, as we expect by approaching a classical transition. The very existence of T^* proves the importance of studying quantum criticality. Although we might be away from the critical coupling, as long as the relation $T < T^*$ holds, we expect to observe the same quantum critical behavior as in Eqs. (14) and (15). Interestingly, T^* plays an important role at $T=0$ as well, the two regions of the scaling function are separated by $2\pi\tau^* = 1/T^*$. This picture is in perfect accord with critical fluctuation close to $y=0$, which change their nature around T^*.

The knowledge of y_t and y_h allows us to formally determine all critical exponents[28] (see Table I). These exponents satisfy the scaling as well as quantum hyperscaling $[2-\alpha = (d+z)\nu]$ relations.

VI. CLASSICAL LIMIT

The high-temperature regime of the quantum system can be regarded as the finite-sized classical counterpart, with sizes smaller than the coherence length. At finite temperatures, the singular contribution to the free energy in y can be obtained from Eq. (7), but it is instructive to follow a different approach. In the high-temperature limit, the harmonic oscillator becomes classical, i.e., Q and P being classical variables in Eq. (1). Then, after tracing out the electronic degrees of freedom, we obtain the partition function for the oscillator as

$$Z = \int dP dQ \exp\left(-\frac{P^2}{2mT} - \frac{m\omega_0^2 y Q^2}{2T} - \frac{VQ}{T}\right) = \frac{2\pi T}{\omega_0\sqrt{y}} \exp\left(\frac{V^2}{2m\omega_0^2 y T}\right), \tag{19}$$

where we integrate the exponential of the classical energy over all of phase space (all possible momenta and positions), and we added a source term VQ. Then, the singular part of the free energy is

$$f_s(T \gg T^*) = \frac{T}{2}\ln(y) - \frac{V^2}{2m\omega_0^2 y}, \tag{20}$$

giving $\alpha=2$ and $\gamma=1$. This accounts for the instability in the purely classical version of Eq. (1) for the oscillator caused by the vanishing of the vibration frequency.

Dissipation has no effect at high T; the instability is caused by the potential renormalization. At this point, we are dealing with a classical harmonic oscillator; therefore $d_{\text{eff}} = 0$, and the critical theory is formally equivalent to that of a "zero-dimensional" Gaussian model.[28] The other exponents are determined as $\beta=-1/2$ and $\delta=-1$, but the ν and η exponents are senseless, since there is no dimension for the spatial or imaginary time dependence. We mention that the criticality of the lattice version of Eq. (1), the Holstein

model, is expected to depend on the dimensionality of the electrons, and belong to a different universality class.

VII. SUMMARY

Motivated by the possibility of reaching WB singularity in carbon nanotubes,[21] we have studied the critical properties of a variant of the WB singularity triggered by local electron-vibration interaction. The only relevant energy scale of the problem is identified as T^*, which separates the quantum and classical critical regions. The former belongs to the universality class of weakly first-order quantum-phase transitions, while the latter is formally equivalent to a "zero-dimensional" Gaussian model. The two qualitatively different critical regimes are accessible by local vibrational spectroscopy[11] and influence the electronic response (e.g., dephasing[8]) as well.

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