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Thermal expansion and Grüneisen parameter in quantum Griffiths phases

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We consider the behavior of the Grüneisen parameter, the ratio between thermal expansion and specific heat, at pressure-tuned infinite-randomness quantum-critical points and in the associated quantum Griffiths phases. We find that the Grüneisen parameter diverges as $\ln(T_0/T)$ with vanishing temperature T in the quantum Griffiths phases. At the infinite-randomness critical point itself, the Grüneisen parameter behaves as $[\ln(T_0/T)]^{1+1/(\nu\psi)}$ where ν and ψ are the correlation length and tunneling exponents. Analogous results hold for the magnetocaloric effect at magnetic-field-tuned transitions. We contrast clean and dirty systems, we discuss subtle differences between Ising and Heisenberg symmetries, and we relate our findings to recent experiments on $CePd_{1-x}Rh_x$.

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I. INTRODUCTION

Quantum phase transitions (QPTs) occur at zero temperature when a parameter such as pressure or magnetic field is varied. At these transitions, the quantum fluctuations associated with the competition between different quantum ground states lead to unconventional thermodynamic and transport properties.[1–4] In metallic systems they can induce, e.g., non-Fermi-liquid behavior and exotic superconductivity. The characterization of QPTs is a topic of great current interest with many fundamental questions remaining unresolved.

Over the last few years, the Grüneisen parameter Γ, the ratio between thermal-expansion coefficient and specific heat, has become a valuable tool for analyzing pressure-tuned QPTs. For transitions tuned by magnetic field, the same role is played by the magnetocaloric effect. Zhu *et al.*[5,6] showed that the thermal-expansion coefficient is more singular than the specific heat at a generic clean quantum-critical point (QCP). They thus predicted that the Grüneisen parameter diverges when approaching criticality. Specifically, if hyperscaling holds (below the upper critical dimension), $\Gamma \sim T^{-1/(z\nu)}$ if the temperature T is lowered at the critical pressure p_c and $\Gamma \sim 1/(p-p_c)$ if the pressure p approaches p_c at zero temperature (z denotes the dynamical exponent). Above the upper critical dimension, Γ still diverges, but the functional form is modified by dangerously irrelevant variables. Diverging Grüneisen parameters have since been observed at several[7–10] magnetic QCPs.

Since many materials feature considerable amounts of quenched randomness, the study of QPTs in random systems has received much attention recently. The interplay between quantum fluctuations and static random fluctuations results in more dramatic disorder effects at QPTs than at classical transitions, including quantum Griffiths singularities,[11–15] activated dynamical scaling,[16–18] and smeared transitions.[19,20] A review of some of these phenomena can be found in Ref. 21. In view of the insight about the character of a QPT that can be gained from the Grüneisen parameter and the magnetocaloric effect, it is desirable to determine their behavior within these unconventional scenarios. This is particularly timely because exotic scaling behavior compatible with many predictions of the quantum Griffiths scenario has recently been observed[22,23] at the ferromagnetic QPT in $CePd_{1-x}Rh_x$.

In this Rapid Communication, we therefore calculate the thermal-expansion coefficient and the Grüneisen parameter (for pressure-tuned transitions), as well as the magnetocaloric effect (for magnetic-field-tuned transitions) at infinite-randomness QCPs and in the associated quantum Griffiths phases. We use two methods: a heuristic rare-region theory and a scaling analysis of the QCP itself.

We define the Grüneisen parameter[5,24] Γ as the ratio between the thermal volume expansion coefficient,

$$\beta = V^{-1}(\partial V/\partial T)_p = -V^{-1}(\partial S/\partial p)_T, \tag{1}$$

and the molar specific heat

$$c_p = TN^{-1}(\partial S/\partial T)_p. \tag{2}$$

Here, V is the volume, N is the particle number, and S denotes the entropy. Thus,

$$\Gamma = \frac{\beta}{c_p} = -\frac{(\partial S/\partial p)_T}{V_m T(\partial S/\partial T)_p}, \tag{3}$$

with $V_m = V/N$ as the molar volume. For a pressure-tuned transition, $(\partial S/\partial p)_T = p_c^{-1}(\partial S/\partial r)_T$ explores the dependence of the entropy on the dimensionless distance from criticality, $r=(p-p_c)/p_c$. For a transition tuned by magnetic field H with $r=(H-H_c)/H_c$, the same dependence is encoded in $(\partial S/\partial H)_T = (\partial M/\partial T)_H$, with M as the total magnetization. We thus define the magnetic analog of the Grüneisen parameter,

$$\Gamma_H = -\frac{(\partial M/\partial T)_H}{c_H} = -\frac{(\partial S/\partial H)_T}{T(\partial S/\partial T)_H} = \frac{1}{T}\left(\frac{\partial T}{\partial H}\right)_S, \tag{4}$$

which can be determined from the magnetocaloric effect.

II. RARE-REGION THEORY

For definiteness, we consider a d-dimensional quantum Landau-Ginzburg-Wilson (LGW) free-energy functional for an n-component order-parameter field ϕ. The action of the clean system is given by[25]

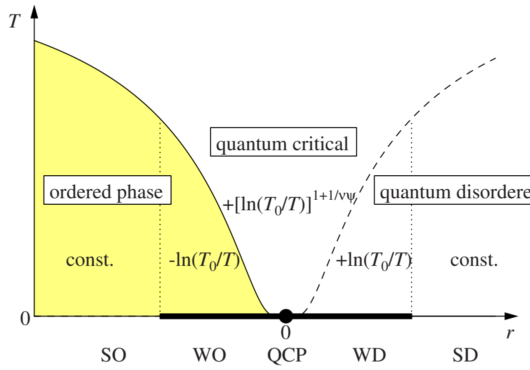

FIG. 1. (Color online) Schematic phase diagram close to an infinite-randomness QCP. SO and SD denote the strongly ordered and disordered bulk phases, while WO and WD are the weakly ordered and disordered quantum Griffiths phases. The terms in the figure show the temperature dependence of Γ or Γ_H (for pressure and field-tuned transitions, respectively).

$$S=\int dxdy\,\phi(x)K(x,y)\phi(y)+u\int dx\,\phi^4(x). \tag{5}$$

Here, $x\equiv(\mathbf{x},\tau)$ comprises position $\mathbf{x}$ and imaginary time τ, and $\int dx\equiv\int d\mathbf{x}\int_0^{1/T}d\tau$. The Fourier transform of the bare inverse propagator (two-point vertex) $K(x,y)$ reads $K(\mathbf{q},\omega_n)=(r_0+\mathbf{q}^2+\gamma|\omega_n|^{2/z_0})$ with r_0 being the bare distance from the (clean) QCP. To introduce quenched randomness, we dilute the system with nonmagnetic impurities of spatial density b; i.e., we add a potential, $\delta r(\mathbf{x})=\Sigma_i V[\mathbf{x}-\mathbf{x}(i)]$, to r_0. Here, $\mathbf{x}(i)$ are the random positions of the impurities and $V(\mathbf{x})$ is a positive short-ranged impurity potential.

In our disordered LGW theory, quantum Griffiths phases occur in Ising systems ($n=1$) with dissipationless dynamics ($z_0=1$) or in continuous-symmetry systems ($n>1$) with Ohmic dissipation ($z_0=2$).[21] We first focus on the Ising case; minor differences for $n>1$ will be discussed later.

We start our analysis with the weakly disordered (WD) quantum Griffiths phase (WD in Fig. 1). Despite the dilution, there are large spatial regions devoid of impurities. They can be locally in the magnetic phase even though the bulk system is still nonmagnetic. The probability w of finding such a rare region or cluster of linear size L_{RR} is exponentially small in its volume, $w\sim\exp(-bL_{RR}^d)$. Because the cluster is locally ordered it acts as a two-level system with an energy gap $\epsilon\sim\exp(-aL_{RR}^d)$. Combining the two exponential laws, we obtain the well-known power-law density of states (see, e.g., Ref. 21),

$$\rho(\epsilon)\sim\epsilon^{\lambda(r)-1}. \tag{6}$$

We have parametrized the nonuniversal power law in terms of the Griffiths exponent $\lambda=b/a$. It vanishes at the QCP and increases with increasing distance r from criticality. To determine the rare-region contribution to the entropy at temperature T, we note that each rare region with $\epsilon<T$ contributes an entropy of $\ln 2$ while those with $\epsilon>T$ do not contribute significantly. Allowing for an r-dependent prefactor, we thus find

$$S(r,T)=Ng(r)(T/T_0)^{\lambda(r)}, \tag{7}$$

where T_0 is a microscopic temperature scale.

Thermal-expansion coefficient and specific heat can now be calculated easily by taking the appropriate derivatives of the entropy. From Eqs. (1) and (2), we find the leading low-temperature behavior to be

$$\beta=\frac{1}{V_m p_c}g(r)\lambda'(r)(T/T_0)^{\lambda(r)}\ln(T_0/T), \tag{8}$$

$$c_p=g(r)\lambda(r)(T/T_0)^{\lambda(r)}, \tag{9}$$

where $\lambda'(r)$ denotes the derivative of λ with respect to r. In the Grüneisen ratio, the temperature dependencies of β and c_p almost completely cancel, resulting in

$$\Gamma=\frac{\beta}{c_p}=\frac{1}{V_m p_c}\frac{\lambda'(r)}{\lambda(r)}\ln(T_0/T). \tag{10}$$

The rare-region contribution to the Grüneisen parameter diverges logarithmically with decreasing temperature in the entire WD quantum Griffiths phase. Because λ increases with r, both the thermal-expansion coefficient and the Grüneisen parameter are positive. This agrees with the notion that the low-temperature entropy decreases with increasing distance from criticality.

In the weakly ordered (WO) quantum Griffiths phase, the relevant degrees of freedom are strongly coupled clusters that are sufficiently isolated from the (ordered) bulk system so that they can fluctuate independently. This requires that the effective coupling J_{eff} of the cluster to the bulk is smaller than its energy gap, which still reads $\epsilon\sim\exp(-aL_{RR}^d)$. To isolate the cluster, it must thus be surrounded by a large spatial region that is locally in the nonmagnetic phase. Generically, the correlations will drop off exponentially with distance in this region. The condition $J_{\text{eff}}<\epsilon$ thus implies that the *linear* size of the isolating region must vary as $\ln(1/\epsilon)\sim L_{RR}^d$ with the cluster size L_{RR}. We conclude that the probability of finding a sufficiently isolated cluster of size L_{RR} drops off as $w\sim\exp[-\bar{b}(L_{RR}^d)^d]$, i.e., much faster than in the WD phase. The resulting density of states takes the form

$$\rho(\epsilon)\sim\frac{1}{\epsilon}\exp[-\bar{\lambda}(r)\ln^d(\epsilon_0/\epsilon)], \tag{11}$$

with a nonuniversal $\bar{\lambda}(r)$ that is the analog of the Griffiths exponent $\lambda(r)$. Thus, we still obtain a gapless spectrum, but the singularity is weaker than in the WD phase in all dimensions $d>1$. In particular, the density of states vanishes faster than any power law with $\epsilon\to 0$. We emphasize that Eq. (11) is the generic result; special types of randomness can lead to stronger singularities. For instance, in a percolation scenario (site or bond dilution of a lattice), a shell of empty sites or bonds is sufficient to completely isolate a cluster. In this case, $w\sim\exp[-\bar{b}L_{RR}^{d-1}]$ and $\rho(\epsilon)\sim\epsilon^{-1}\exp[-\bar{\lambda}(r)\ln^{1-1/d}(\epsilon_0/\epsilon)]$ giving rise to a singularity even stronger than in the WD phase.[26]

The rare-region contribution to the entropy can be estimated as above by simply counting the clusters with energy gap $\epsilon < T$. This gives

$$S(r,T) = N\bar{g}(r)\exp[-\bar{\lambda}(r)\ln^d(T_0/T)]. \quad (12)$$

Using Eqs. (1) and (2), we obtain the leading low-temperature behavior of thermal expansion and specific heat,

$$\beta = \frac{\bar{g}(r)\bar{\lambda}'(r)}{V_m p_c}\exp[-\bar{\lambda}(r)\ln^d(T_0/T)]\ln^d(T_0/T), \quad (13)$$

$$c_p = d\bar{g}(r)\bar{\lambda}(r)\exp[-\bar{\lambda}(r)\ln^d(T_0/T)]\ln^{d-1}(T_0/T). \quad (14)$$

This results in a Grüneisen parameter of

$$\Gamma = \frac{\beta}{c_p} = \frac{1}{dV_m p_c}\frac{\bar{\lambda}'(r)}{\bar{\lambda}(r)}\ln(T_0/T), \quad (15)$$

which is (except for the extra factor $1/d$) identical to the WD result [Eq. (10)]. Note that a different power of $\ln(\epsilon_0/\epsilon)$ in the exponent of Eq. (11) (as discussed above for percolation disorder) would not change the temperature dependence of Γ. In the WO quantum Griffiths phase, $\bar{\lambda}$ decreases with increasing r (approaching the QCP). The thermal-expansion coefficient and the Grüneisen parameter are therefore negative, again in agreement with the entropy decreasing with increasing distance from criticality. Note that the rare-region contributions to both β and c_p in the WO phase are only weakly singular for $d>1$. Therefore, they may be subleading to contributions from other soft modes in the system, making Eq. (15) hard to observe experimentally.

III. SCALING ANALYSIS

We now complement the heuristic rare-region theory by a scaling analysis of the Grüneisen parameter at infinite-randomness QCPs. These exotic critical points emerge from the strong-disorder renormalization group[16,27,28] and generally occur in conjunction with the quantum Griffiths phases discussed above.[21]

According to the strong-disorder renormalization group, the density of independent clusters surviving at temperature T scales such as an inverse volume and thus has the scaling form[29]

$$n(r,T) = [\ln(T_0/T)]^{-d/\psi}\Phi(r[\ln(T_0/T)]^{1/(\nu\psi)}), \quad (16)$$

where ν and ψ are the correlation length and tunneling exponents. The scaling function $\Phi(y)$ is analytic at $y \geq 0$ (because there is no finite-temperature phase transition at $r \geq 0$). For small y, we can thus expand $\Phi(y) = \Phi(0) + y\Phi'(0) + \cdots$. For large positive y (in the WD quantum Griffiths phase), $\Phi(y) = Ay^{d\nu}\exp(-cx^{\nu\psi})$ with A and c constants. The scaling function $\Phi(y)$ has a singularity at some $y_c < 0$ marking the transition to the ordered phase. This immediately gives the unusual form of the phase boundary, $T_c(r) = T_0\exp[-(y_c/r)^{\nu\psi}]$, sketched in Fig. 1. Since the surviving clusters are essentially free, each contributes $s_0 = \ln 2$ to the entropy. The scaling part of the entropy thus reads as

$$S(r,T) = Ns_0[\ln(T_0/T)]^{-d/\psi}\Phi(r[\ln(T_0/T)]^{1/(\nu\psi)}). \quad (17)$$

We first calculate the thermal-expansion coefficient and the specific heat at criticality, $r=0$. Applying Eqs. (1) and (2) to scaling form (17) of the entropy, we obtain

$$\beta = -\frac{s_0}{V_m p_c}\Phi'(0)[\ln(T_0/T)]^{-d/\psi + 1/(\nu\psi)}, \quad (18)$$

$$c_p = \frac{s_0 d}{\psi}\Phi(0)[\ln(T_0/T)]^{-d/\psi - 1}. \quad (19)$$

Forming the ratio β/c_p, we find that the critical part of the Grüneisen parameter behaves as

$$\Gamma = -\frac{\psi}{V_m p_c d}\frac{\Phi'(0)}{\Phi(0)}[\ln(T_0/T)]^{1+1/(\nu\psi)}. \quad (20)$$

Equation (20) holds in the entire quantum-critical region $T > T_0\exp[-|y_x/r|^{\nu\psi}]$ where the constant y_x marks the crossover of $\Phi(y)$. The sign of Γ does not follow from the scaling analysis, but because the entropy accumulates close to the finite-temperature phase boundary, we generally expect $\Gamma > 0$ in the quantum-critical region.[6]

Let us now analyze scaling form (17) of the entropy in the WD quantum Griffiths phase, i.e., for $r>0$ and $T < T_0\exp[-(y_x/r)^{\nu\psi}]$. Using the large-argument limit of the scaling function $\Phi(y)$, the density of surviving clusters is given by $n(r,T) = Ar^{d\nu}\exp[-cr^{\nu\psi}\ln(T_0/T)]$. The resulting functional form of the entropy,

$$S(r,T) = Ng(r)(T/T_0)^{\lambda(r)}, \quad (21)$$

is identical to that found in Eq. (7) using heuristic rare-region arguments, but the scaling analysis also gives $\lambda(r) = cr^{\nu\psi}$ and $g(r) = As_0 r^{d\nu}$ in terms of the distance to criticality and the critical exponents. Inserting $g(r)$ and $\lambda(r)$ into Eqs. (8)–(10) leads to

$$\beta = \frac{As_0}{V_m p_c}r^{d\nu + \nu\psi - 1}c\nu\psi(T/T_0)^{\lambda(r)}\ln(T_0/T), \quad (22)$$

$$c_p = As_0 c r^{d\nu + \nu\psi}(T/T_0)^{\lambda(r)}, \quad (23)$$

$$\Gamma = \frac{1}{V_m}\frac{\nu\psi}{p - p_c}\ln(T_0/T). \quad (24)$$

The prefactor of the logarithmic temperature dependence of Γ thus diverges as $1/(p-p_c)$ at the QCP.

As discussed above, the behavior on the ordered side of the transition, i.e., in the WO quantum Griffiths phase, is dimensionality and disorder dependent. Once these are fixed, the analysis can be performed in complete analogy to the WD quantum Griffiths phase.

IV. CONCLUSIONS

We have determined the Grüneisen parameter Γ at pressure-tuned QPTs in the presence of quenched disorder. At an infinite-randomness QCP, the critical contribution to Γ diverges as $[\ln(T_0/T)]^{1+1/(\nu\psi)}$ with $T \to 0$. In the associated quantum Griffiths phases on both sides of the QCP, the rare-region contribution to Γ behaves as $\ln(T_0/T)$ with a prefactor that diverges and changes sign at criticality ($\Gamma<0$ for $p<p_c$ and $\Gamma>0$ for $p>p_c$). Our results must be contrasted with the behavior at clean QCPs, where Γ diverges as a power of T at criticality but remains finite for all $p\neq p_c$.

In many systems, a QPT can be induced by doping instead of pressure. If the main effect of doping is an expansion or compression of the lattice, it acts as "chemical pressure." Close to criticality, the effects of pressure p and doping x can then be related via $(p-p_c)=c(x-x_c)$ with c a constant. Defining the distance from criticality for such a transition as $r=(x-x_c)/x_c$, this leads to the relation $\partial/\partial p=(cx_c)^{-1}\partial/\partial r$. All our results for β and Γ thus hold if cx_c is substituted for p_c.

If the transition is tuned by magnetic field instead of pressure, our calculations carry over to the magnetocaloric effect Γ_H defined in Eq. (4). In fact, by replacing $V_m p_c$ by H_c in the results for Γ, one obtains the corresponding expressions for Γ_H. Note, however, that our analysis assumes random-T_c-type disorder that does not break the order-parameter symmetry. In magnetic-field-tuned transitions in the presence of disorder, stronger random-field-type effects may be generated.[30–33] They would require a separate analysis.

In our LGW theory, quantum Griffiths phases and infinite-randomness QCPs occur either for Ising symmetry without dissipation or for continuous $O(n)$ symmetry ($n>1$) and Ohmic dissipation.[21] So far, we have focused on the Ising case. The main difference for $n>1$ is that ordered clusters act as (damped) quantum rotors rather than two-level systems.[29] In the Griffiths phase, this changes the prefactors $g(r)$ and $\bar{g}(r)$ while the exponents $\lambda(r)$ and $\bar{\lambda}(r)$ remain the same. At criticality, entropy (17) picks up an extra factor $\ln(T_0/T)$ from the entropy of a single rotor. It drops out in the ratio β/c_p. Thus, our results remain valid in the $O(n)$ case, at least at criticality and in the WD Griffiths phase. In the WO Griffiths phase, the rare-region contributions to β and c_p will be overcome by conventional soft-mode terms [because the dissipative $O(n)$ system is gapless].

We now turn to experiment. $CePd_{1-x}Rh_x$ features a ferromagnetic QPT that appears to be dominated by rare regions.[22,23] The phase boundary develops a tail characteristic of a smeared QPT; and at temperatures above the tail, magnetization, susceptibility, and specific heat display nonuniversal power laws as expected in a quantum Griffiths phase. Recent measurements of the thermal expansion[23] resulted in a very weakly temperature-dependent Grüneisen parameter close to the putative transition at $x_c\approx 0.87$, in qualitative agreement with our theory. However, the variation in Γ with doping x differs considerably from our results. This may be caused by the fact that the doping is not isoelectronic. It thus not only acts as chemical pressure by inducing a lattice compression (as assumed in our discussion) but it also changes the electronic structure directly. To disentangle these effects one could prepare a sample with doping close to x_c and then drive it through the QPT by pressure.

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