$T = 0$ Heavy-Fermion Quantum Critical Point as an Orbital-Selective Mott Transition

Lorenzo De Leo,[1,2] Marcello Civelli,[3] and Gabriel Kotliar[2]

[1]*Centre de Physique Théorique, Ecole Polytechnique, CNRS, 91128 Palaiseau, France*
[2]*Department of Physics, Rutgers University, Piscataway, New Jersey 08854, USA*
[3]*Theory Group, Institut Laue Langevin, 38042 Grenoble Cedex, France*

(Received 20 April 2008; published 19 December 2008)

We describe the $T = 0$ quantum phase transition in heavy-fermion systems as an orbital-selective Mott transition (OSMT) using a cluster extension of dynamical mean-field theory. This transition is characterized by the emergence of a new intermediate energy scale corresponding to the opening of a pseudogap and the vanishing of the low-energy hybridization between light and heavy electrons. We identify the fingerprint of Mott physics in heavy electron systems with the appearance of surfaces in momentum space where the self-energy diverges and we derive experimental consequences of this scenario for photoemission, compressibility, optical conductivity, susceptibility, and specific heat.

DOI: 10.1103/PhysRevLett.101.256404 PACS numbers: 71.27.+a, 71.10.Hf, 75.20.Hr, 75.30.Mb

Heavy-fermion materials containing electrons in open $4f$ or $5f$ shells and in broad spd bands, continue to be a subject of great interest in condensed matter physics [1]. The description of the antiferromagnetic-paramagnetic transition in these systems is highly nontrivial, because, in addition to the fluctuations of the magnetic order parameter, one has to take into account the changing character, from itinerant to localized, of the f electrons [2,3]. Recent publications [4,5] have debated whether the quantum phase transitions observed in heavy fermions can be described as an orbital-selective Mott transition (OSMT), i.e., a Mott transition taking place in the f orbitals with the spd orbitals remaining itinerant. Some finite temperature aspects of this phenomena are captured by *single-site* dynamical mean-field theory (DMFT) [4,6,7], a method that captures well the peculiar dynamics of the Mott transition assuming a purely local approximation. In Refs. [5,8,9] however, the authors have shown by means of slave boson techniques that going beyond the local approximation is an essential ingredient to obtain an OSMT at zero temperature.

In this Letter we overcome the spatial limitation of DMFT by using one of its cluster extensions (the cellular DMFT, CDMFT [10]), which takes into account short-ranged correlation, and we demonstrate the existence of a $T = 0$ OSMT. In Ref. [11], we have fully characterized the phase diagram of a heavy-fermion model across a quantum critical phase transition, separating a strongly renormalized Fermi liquid from an antiferromagnetic phase. Here, by constraining the mean-field nonordered solution, we focus on the qualitative evolution of the electronic structure. In this way, we isolate the physics that stems directly from the localization of the f electrons from the physics of the magnetic order that intervenes at low temperature in a given material.

We show, in particular, that at the transition a new energy scale emerges. In this energy range a pseudogap opens in the f spectra and the hybridization between heavy f and light spd electrons goes to zero, leading to a complete decoupling of the two bands. Beyond this energy range the f-spd hybridization remains finite. These phenomena have a clear interpretation in terms of an OSMT, revealed by the appearance of surfaces of diverging self-energy in momentum space, fingerprint of a Mott mechanism (Mottness [12]). In the conclusions, we derive a set of experimental consequences relevant for the normal state of real materials close to the quantum critical point at temperatures above the ordered state.

We study the quantum phase transition driven by a hybridizing parameter V in the periodic Anderson model, which describes free spd electrons locally hybridized to nondispersing strongly correlated f electrons. The Hamiltonian is

$$H = \sum_{\mathbf{k}} (\varepsilon_{\mathbf{k}} - \mu) d^{\dagger}_{\mathbf{k}\sigma} d_{\mathbf{k}\sigma} + V \sum_{\mathbf{k}} (f^{\dagger}_{\mathbf{k}\sigma} d_{\mathbf{k}\sigma} + \text{H.c.}) + (E_f - \mu) \sum_{\mathbf{k}} f^{\dagger}_{\mathbf{k}\sigma} f_{\mathbf{k}\sigma} + U \sum_{i} f^{\dagger}_{i\uparrow} f_{i\uparrow} f^{\dagger}_{i\downarrow} f_{i\downarrow}, \quad (1)$$

where $d^{\dagger}_{\mathbf{k}\sigma}$ [$f^{\dagger}_{\mathbf{k}\sigma}$] creates an spd [f] electron with momentum $\mathbf{k}$ and spin σ. The conduction band dispersion is $\varepsilon_{\mathbf{k}} = -\frac{1}{3}(\cos k_x + \cos k_y + \cos k_z)$, the other parameters $U = 10$, $\mu = 0.2$ and $E_f - \mu = -5.7$. The spd and f electrons Green's functions can be written in terms of the f electron self-energy Σ:

$$G_{\alpha}(\omega, \mathbf{k}) = \left[\omega + X_{\alpha}(\mathbf{k}, \omega) - \frac{V^2}{\omega + Y_{\alpha}(\mathbf{k}, \omega)} \right]^{-1}, \quad (2)$$

where $\alpha = f, \quad spd, \quad X_f(\mathbf{k}, \omega) = \mu - E_f - \Sigma(\mathbf{k}, \omega)$, $Y_f(\mathbf{k}, \omega) = \mu - \varepsilon_{\mathbf{k}}$ and $X_{spd} = Y_f$, $Y_{spd} = X_f$.

We implement CDMFT on a two-site cluster [11]. We believe this is the minimal unit able to capture the physics close to the transition point. The Hamiltonian in Eq. (1) is mapped onto an effective two impurity Anderson model

(2IAM) and solved self-consistently via the Lanczos method [13], which introduces a finite energy resolution on the Matsubara axis [14] $\omega_n = (2n-1)\pi/\beta$, with $\beta = 100$.

In order to physically interpret our results, we extract the momentum dependent lattice self-energy $\Sigma(\mathbf{k},\omega)$ in Eq. (2) from the cluster quantities, restoring the cubic lattice symmetry. Various methods have been proposed in the literature [14]. Close to the transition, where particles tend to localize, it has been shown [15] that a suitable quantity to adopt is the cluster cumulant $\hat{M}(\omega) = [(\omega + \mu - E_f)\hat{\mathbf{1}} - \hat{\Sigma}(\omega)]^{-1}$. In our case we have $M(\mathbf{k}) = M_0 + \frac{1}{3}M_1(\cos k_x + \cos k_y + \cos k_z)$ where $M_0 = M_{11} = M_{22}$ and $M_1 = M_{12} = M_{21}$, and $\Sigma(\mathbf{k},\omega) = \omega + \mu - E_f - M(\mathbf{k},\omega)^{-1}$. A stringent self-consistent test of this periodization can be obtained by recalculating the local f electron Green's function $\sum_{\mathbf{k}} G_f(\mathbf{k},\omega)$, via Eq. (2), and confronting it with the cluster counterpart, direct output of the CDMFT calculation. In Fig. 1 we show the low-energy imaginary parts of the local f Green's functions (the density of states DOS). The good agreement between the periodized f DOS and the cluster f DOS validates our procedure. Moreover, we show the DOS for the spd electrons and the effective hybridization $\mathrm{Re}G_{f-spd} = \sum_{\mathbf{k}} \mathrm{Re}G_{f-spd}(\omega,\mathbf{k})$ [6]. These quantities demonstrate that, as a function of the tuning parameter V, the system undergoes a phase transition. The numerical uncertainties become greater near the transition, hence we cannot determine whether the transition is second order, as predicted in a scaling theory [16] (in which case the best fit of the pseudogap scaling with a power law gives an exponent $z\nu \sim 0.33$), or first order as found in a recent Guzwiller treatment [3]. For $V > V^* \sim 0.58$ the system is in the heavy-fermion phase where the f electrons present a Kondo peak at the Fermi level $\omega = 0$ and take active part in the conduction. The strong hybridization with the spd electrons is evident in the suppression of the spd DOS and in the nonzero value of the effective hybridization $f - spd$ close to $\omega = 0$. The intensity of the f peak reduces approaching V^*, while at the same time the spd spectral weight enhances. For $V < V^*$ the system is in an orbital-selective Mott state where the f electron spectrum has a gap. The f electron spectral weight is not completely transferred from low energy to the Hubbard bands (placed around $\omega \sim \pm 5$) but rather to a new intermediate energy scale, giving rise to a pseudogap. Within this pseudogap the spd electrons recover the free band DOS and the effective hybridization is zero. This shows that the spd band at low energy is completely decoupled from the f band, but the effective hybridization remains active at a finite intermediate energy scale.

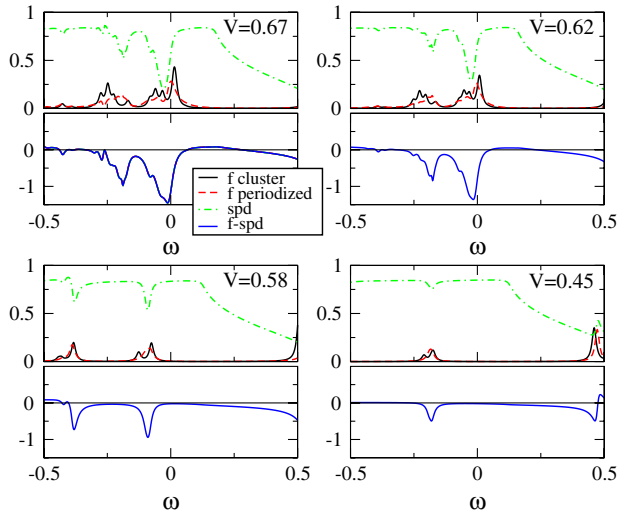

FIG. 1 (color online). The local density of states (DOS) as a function of the hybridization V in a low-energy window around the chemical potential (Hubbard bands are out of the picture at $\omega \sim \pm 5$). Black continuous lines are the cluster f-electron DOS. The red dashed (green dot-dashed) lines are the $f[spd]$-electron DOS $-\frac{1}{\pi}\sum_{\mathbf{k}} G_{f[spd]}(\mathbf{k},\omega)$. The bottom panels show the effective hybridization $\sum_{\mathbf{k}} \mathrm{Re}G_{f-spd}(\omega,\mathbf{k})$ (blue continuous line).

We can now display the quasiparticle bands along some specific cuts in the $\omega - \mathbf{k}$ space. From Eq. (2), we notice that G_f transforms into G_{spd} upon the exchange of $E_f - \Sigma(\mathbf{k},\omega)$ with $\varepsilon_{\mathbf{k}}$. The poles of the f and spd Green's function are therefore the same, provided the $\mathrm{Im}\Sigma_{\mathbf{k}}$ is small as in Fermi liquid theory, and the same resolving equation is obtained for either $\alpha = f, spd$:

$$\omega + \mu - E_f - \mathrm{Re}\Sigma(\mathbf{k},\omega) = V^2/(\omega + \mu - \varepsilon_{\mathbf{k}}). \quad (3)$$

The spectral-weight contribution to the electronic bands coming from f and spd electrons are, however, very different. In Fig. 2 we show the f (top row) and spd (bottom row) spectral functions $-\frac{1}{\pi}\mathrm{Im}G_{f[spd]}(\mathbf{k},\omega)$ along the path $X = (\pi,0,\pi) \to \Gamma = (0,0,0) \to \Pi = (\pi,\pi,\pi)$ of momentum space, for varying hybridization-parameter V

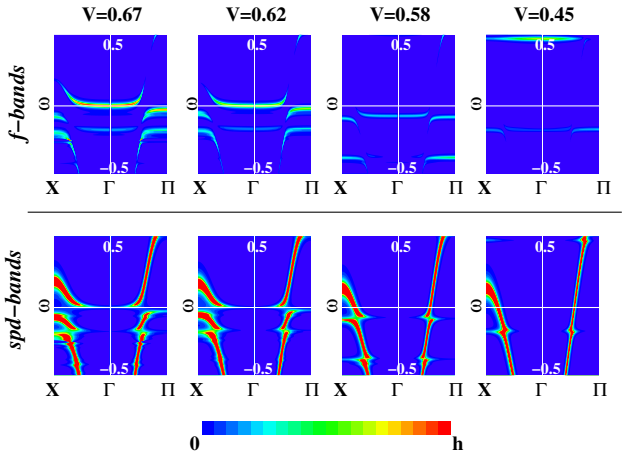

FIG. 2 (color online). Evolution across the transition point ($V^* \sim 0.58$) of the f- (top row) and spd- (bottom row) electron spectral functions along the path $X = (\pi,0,\pi) \to \Gamma = (0,0,0) \to \Pi = (\pi,\pi,\pi)$ in momentum space. The color scale (bottom legend) is $h = 1.0[2.0]$ for $f[spd]$ electrons.

(from left to right). For $V = 0.67 > V^*$ the band crossing the Fermi level has predominantly f character at low energy and a strongly renormalized effective mass. In approaching the transition point the f electron contribution quickly reduces until disappearing completely from the Fermi level for $V \sim 0.58 = V^*$. In addition, beyond the transition the f band shifts to negative energies. In describing the localization of the f electrons therefore, the double effect of suppression and translation of the f band has to be taken into account. Our result is a prediction that can be observed in photoemission experiments. At the same time, in crossing the transition, the effective mass of the spd electrons is reduced to the free value.

Recent studies [15] have shown that insights into quantum phase transition phenomena can be attained by studying not only the Fermi surface (i.e., poles of the Green's function), but also surfaces of zeroes in the Green's functions (i.e., poles of the self-energy). We first remark that in our model there is always a $G_f = 0$ surface in momentum space, corresponding to the free conduction electron Fermi surface FS0 (given by $\varepsilon_{\mathbf{k}} - \mu = 0$). Further surfaces of zeroes ZS in G_f can appear in $\mathbf{k}$-space if there are $\mathbf{k}$ points for which $\Sigma(\vec{k}, 0) \rightarrow \infty$. In this case we observe that G_{spd} reduces to the free conduction electron Green's function [Eq. (2)]. We show that this latter phenomenon indeed takes place in approaching the transition point V^*. In Fig. 3 we present the Fermi Surface FS [determined by $\omega \rightarrow 0$ in Eq. (3)], FS0 and ZS for different values of the hybridizing coupling V across the transition point. For convenience sake, only the lower half of the three-dimensional Brillouin zone is shown. In the heavy-fermion phase $V = 0.67 > V^*$, only FS and FS0 are visible at $\omega = 0$ and far apart in momentum space. In this case FS0 is not relevant for the low-energy physics of the system. As soon as $V \lesssim V^*$, however, a small ZS appears around the point $\mathbf{k} = (0, 0, 0)$ which pushes FS to collapse onto the free FS0. Since at FS0 $G_f \rightarrow 0$, this effect originates the strong suppression and disappearance of the f spectral weight at the Fermi level (see Fig. 2). The appearance of ZS can be already seen in the cluster quantities, which are displayed in the $V - i\omega_n$ space in the bottom of Fig. 3. At $V = V^*$ a divergence takes place for $\omega_n \rightarrow 0$ [11] in the even eigenvalue of the cluster self-energy Σ_{ev} (left panel), which, via the periodization procedure, corresponds to the lattice self-energy at $\mathbf{k} = (0, 0, 0)$. By further reducing V below V^*, ZS travels from $\mathbf{k} = (0, 0, 0)$ to $\mathbf{k} = (\pi, \pi, -\pi)$, where the divergence appears in Σ_{odd} for $V = 0.43$ (right panel). The position in $\mathbf{k}$-space of the FS remains unchanged for $V < 0.58$, numerically overlapping with FS0. This indicates that at the Fermi level $G_f = 0$, i.e., the f electrons remain in a Mott state, and G_{spd} reduces to the free Green's function.

The appearance of a divergent self-energy proves that Mottness is the physical mechanism governing the localization of f-electrons. In an OSMT not all orbitals undergo a localization. In the metallic phase all the orbitals participate in determining the Fermi volume, but, after the transition took place, some ''selected'' orbitals do not contribute to the Luttinger counting anymore. Across this transition a change in the compressibility of the system is expected as localized orbitals become incompressible. This is observed in the actinide series, where the Mott transition can be driven, e.g., by pressure [17]. A Mott transition is also characterized by a significant rearrangement of the electronic structure, since there is a transfer of spectral weight from low to high energies. In our case the spectral weight is not entirely transferred from the Fermi level to the Hubbard bands, but to an intermediate energy scale giving rise to a pseudogap. The resulting modifications of the quasiparticle dispersion can be understood in terms of divergence of the self-energy similarly, e.g., to the pseudogap of cuprates (see Ref. [15]). Hence, in general, both a Fermi volume change and a significant rearrangement of the bands are expected when a material undergoes a Mott transition. Experimentally this would be detected by jumps in the Hall coefficient [18] and in the de Haas–van Alphen frequencies [19]. Measuring the phonon dispersions as a function of temperature is another powerful probe of the orbitally selective Mott transition, as suggested in Ref. [20]. Furthermore the analysis of the 2IAM underlying our self-consistent solution [11] suggests other experimental predictions. Close to the transition the particle-hole symmetry breaking in the lattice model generates a leading irrelevant operator which is forbidden in the sym-

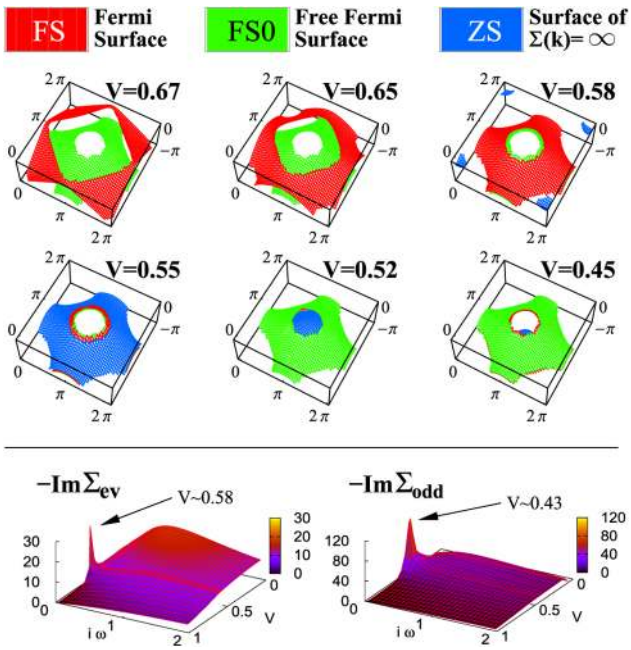

FIG. 3 (color online). Evolution of the Fermi Surface (FS, red) and the surface of diverging self-energy (ZS, blue) across the transition point V^*. An important role is played by the free conduction electron Fermi Surface (FS0, green). The Mott character of the transition is marked by the divergence of the self-energy, detected by the cluster self-energy eigenvalues Σ_{ev} at $V \sim 0.58$ and Σ_{odd} at $V \sim 0.43$, as shown in the $V - i\omega_n$ space (bottom panel).

metric case and that causes a log$T$ divergence in the specific heat coefficient at high temperature [21], together with a log$T$ divergence in the staggered spin susceptibility and in the pairing susceptibility. On the other hand, the formation of singlet correlations on the energy scale of the pseudogap in the Mott-insulating phase implies a depression of the uniform spin susceptibility at low temperature which has been observed, for example, in $CeRhIn_5$ but not in $CeCoIn_5$ [22,23]. Since these effects originate from the competition of Kondo screening and RKKY interaction, we expect them to take place below a temperature comparable to (the largest of) these two energy scales (roughly 10^{-3} the bandwidth, corresponding approximately to 10 K in $CeRh[Co]In_5$). This can be understood considering that $CeRhIn_5$, unlike $CeCoIn_5$, is antiferromagnetic at zero temperature and hence lies on the Mott selective side of the transition. Optical conductivity (not shown) displays a clear hybridization gap in the delocalized phase which is absent in the phase where the f electrons are localized. This is consistent with the experimental assignment of $CeRhIn_5$ to the localized side and $CeCoIn_5$ to the itinerant side of the transition [24].

To summarize, we have discussed the evolution of the momentum resolved spectra of the periodic Anderson model across the quantum phase transition, showing its orbital-selective Mott character. We have described how the electronic structure undergoes dramatic reconstruction: at the transition the f spectral weight is completely suppressed at the chemical potential and a new energy scale emerges in the form of an f-electron pseudogap. Within this latter energy range, the spd band reduces to the free band. f and spd electrons become totally decoupled at low-energy while retaining a finite hybridization at higher energies. We have finally shown that the concept of a surface of diverging self-energy is useful for the understanding of this phenomenon.

It is important to stress that the orbital-selective Mott phase studied here is not a stable phase at $T = 0$ because the f electrons order magnetically as soon as they decouple at low energy from the conduction band [11]. In our calculations the magnetic ordering originates from an instability of the orbital-selective Mott state and not from an instability of the itinerant paramagnetic heavy-fermion state. This supports the interpretation of the magnetic transition as a byproduct of the OSMT (see also Ref. [3]). Our CDMFT study improves previous DMFT studies [4], where the local character of the theory forbids the $T = 0$ OSMT, and previous slave boson studies [5,8,25,26], where a finite bandwidth in the f electrons must be introduced in order to have an exchange mechanism that is not killed by the vanishing of the effective hybridization. In our case such a term is not needed, because retaining the full frequency dependence of the self-energy allows to have a vanishing effective hybridization at the Fermi level, but at the same time an exchange mechanism generated by the nonvanishing hybridization at finite frequency.

We acknowledge useful discussions with M. Fabrizio, A. Georges, C. Pepin, J. Paglione, and I. Paul. This work was supported by NSF Grant DMR 0528969 and ICAM.

[1] H. v. Löhneysen, A. Rosch, M. Vojta, and P. Wölfle, Rev. Mod. Phys. **79**, 1015 (2007); P. Gegenwart, Q. Si, and F. Steglich, Nature Phys. **4**, 186 (2008).
[2] B. Johansson, Philos. Mag. **30**, 469 (1974); J. W. Allen and R. M. Martin, Phys. Rev. Lett. **49**, 1106 (1982); M. Lavagna, C. Lacroix, and M. Cyrot, Phys. Lett. A **90**, 210 (1982);H. Watanabe and M. Ogata, Phys. Rev. Lett. **99**, 136401 (2007).
[3] N. Lanatà, P. Barone, and M. Fabrizio, Phys. Rev. B **78**, 155127 (2008).
[4] L. de' Medici, A. Georges, G. Kotliar, and S. Biermann, Phys. Rev. Lett. **95**, 066402 (2005).
[5] C. Pépin, Phys. Rev. Lett. **98**, 206401 (2007).
[6] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Rev. Mod. Phys. **68**, 13 (1996).
[7] K. Held, A. K. McMahan, and R. T. Scalettar, Phys. Rev. Lett. **87**, 276404 (2001); M. B. Zölfl *et al.*, Phys. Rev. Lett. **87**, 276403 (2001); K. Haule, V. Oudovenko, S. Y. Savrasov, and G. Kotliar, Phys. Rev. Lett. **94**, 036401 (2005).
[8] I. Paul, C. Pépin, and M. R. Norman, Phys. Rev. Lett. **98**, 026402 (2007).
[9] C. Pépin, Phys. Rev. B **77**, 245129 (2008).
[10] G. Kotliar, S. Y. Savrasov, G. Pálsson, and G. Biroli, Phys. Rev. Lett. **87**, 186401 (2001).
[11] L. De Leo, M. Civelli, and G. Kotliar, Phys. Rev. B **77**, 075107 (2008).
[12] F. H. L. Essler and A. M. Tsvelik, Phys. Rev. B **65**, 115117 (2002); I. E. Dzyaloshinski, Phys. Rev. B **68**, 085113 (2003); T. D. Stanescu, P. W. Phillips, and Ting-Pong Choy, Phys. Rev. B **75**, 104503 (2007).
[13] M. Caffarel and W. Krauth, Phys. Rev. Lett. **72**, 1545 (1994).
[14] G. Kotliar *et al.*, Rev. Mod. Phys. **78**, 865 (2006).
[15] T. D. Stanescu and G. Kotliar, Phys. Rev. B **74**, 125110 (2006); T. D. Stanescu, M. Civelli, and G. Kotliar, Ann. Phys. (Leipzig) **321**, 1682 (2006).
[16] M. A. Continentino, Braz. J. Phys. **35**, 197 (2005).
[17] S. Heathman *et al.*, Phys. Rev. Lett. **85**, 2961 (2000).
[18] S. Paschen *et al.*, Nature (London) **432**, 881 (2004).
[19] H. Shishido, R. Settai, H. Harima, and Y. O-nuki, J. Phys. Soc. Jpn. **74**, 1103 (2005).
[20] C. Falter, T. Bauer, and F. Schnetgöke, Phys. Rev. B **73**, 224502 (2006).
[21] I. Affleck, A. W. W. Ludwig, and B. A. Jones, Phys. Rev. B **52**, 9528 (1995).
[22] V. S. Zapf *et al.*, Phys. Rev. B **65**, 014506 (2001).
[23] J. Paglione (private communication).
[24] K. S. Burch *et al.*, Phys. Rev. B **75**, 054523 (2007).
[25] P. Coleman, J. B. Marston, and A. J. Schofield, Phys. Rev. B. **72**, 245111 (2005).
[26] T. Senthil, M. Vojta, and S. Sachdev, Phys. Rev. B **69**, 035111 (2004).