Activation gap in the specific heat measurements for ^{3}He bilayers

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Recently, attention has been given to a system of ^{3}He bilayers where a quantum criticality similar to the one in heavy-fermion compounds has been observed [Neumann *et al.*, Science **317**, 1356 (2007)]. In our previous analysis [Benlagra and Pépin, Phys. Rev. Lett. **79**, 045112 (2007)], based on the Kondo breakdown scenario, we addressed successfully most of the features observed in that experiment. Here, we consider the activation energy Δ observed experimentally in the specific heat measurements at low temperatures in the heavy-Fermi-liquid phase. Within our previous study of this system, this is identified with the gap opening when the upper hybridized band is emptied due to a strong hybridization between the nearly localized first layer and the fluid second one. We discuss the successes and limitations of our approach. An additional prediction is proposed.

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In recent years, the peculiar and nonstandard properties observed in heavy-fermion compounds[1,2] (HFCs) have been the subject of an intense debate. In particular, the nature of the quantum critical point (QCP), thought to be the origin of their remarkable properties in a large region of the phase diagram, remains a puzzling and open problem. Many scenarios have been put forward to account for these properties.[3–8] However, the difficulty with HFCs is their complex chemical structure and the variety of physical phases, often competing, present in their phase diagram. These, despite a growing body of experimental facts, often prevent a ruling in favor of one theoretical model or another.

Lately, a quantum criticality similar to the one in HFCs has been observed in a much simpler system: ^{3}He bilayers adsorbed on a composite substrate of graphite preplated with two solidified layers of ^{4}He.[9] With increase in the total coverage N of ^{3}He atoms, the system seems to undergo a quantum transition, at a critical coverage $N_{\rm crit} = 9.95$ nm^{-2}, separating a regime where the two layers are hybridized and form a bifluid with heavy-Fermi-liquid (FL) properties from a regime where they are completely decoupled, the first one (L1) forming a frustrated two-dimensional (2D) magnet and the second one (L2) remaining fluid. At the putative QCP, there is a breakdown of the FL showing as an apparent divergence of the effective mass m^* and a vanishing of an energy scale T_0 indicative of an effective coupling between the two layers. This is reminiscent of what happens with HFCs except that, in this particular case, there is no long-range magnetic order due to frustration and the energy scales are different.

This similarity has led two of the authors[10] to use one of the models proposed to describe quantum criticality in HFCs, namely, the so-called Kondo breakdown (KB) scenario,[7,8] to account for the remarkable features of the experiment. In that study, the ^{3}He bilayer system has been mapped onto an extended version of the periodic Anderson lattice model, where the nearly localized fermions of L1 are identified with the f electrons and those of L2 with the itinerant conduction electrons. Remarkably, our study identifies the real QCP as occurring at a coverage of $N_I \approx 9.2$ nm^{-2} lower than the experimental one, whereas the experimental QCP is identified as the extrapolation to lower temperatures of a high-energy regime of fluctuations. The coverage $N_I \approx 9.2$ nm^{-2} corresponds to the abrupt increase of the magnetic susceptibility. The QCP in our theory corresponds to an orbital-selective Mott transition at which an effective hybridization between spinons in L1 and conduction electrons in L2 vanishes, resulting in a solidification of L1. It has also been called the Kondo breakdown QCP. This approach was successful enough, using a small set of three fitting parameters, to account for most of the properties observed experimentally. In particular, the apparent presence of two QCPs and the apparent absence of a critical regime in temperature have been explained. The coverage dependence of both the effective mass and the energy scale T_0 have been fitted in an intermediate regime of temperature. We emphasize that the mechanism invoked in our study is different from the one describing the increase of m^* in the second ^{3}He monolayer on bare graphite or pure 2D ^{3}He.[11] The latter indeed does not rely on the interlayer coupling as a crucial aspect of the physics leading to the enhancement of the effective mass.

In this paper, we consider two important additions to our previous theory. First we evaluate the specific heat contribution of the gapped band and compare it to the experimental data. This allows us, in particular, to give a very simple interpretation for the activation gap observed experimentally.[9] The presence of an excitation gap is needed to fit the specific heat measurements in the hybridized phase according to $C_{\rm expt} = \gamma T + \gamma_2 e^{\frac{-\Delta_{\rm expt}}{T}}$, where the first term is usual for a FL with γ proportional to the effective mass of the quasiparticles. The second term defines the experimental activation gap $\Delta_{\rm expt}$. Within our theory, the gap originates from an excitonic mechanism between the heavy and the light bands of the system. The gap Δ is shown to open (see Fig. 1) when the upper hybridized band is emptied due to a strong hybridization between the nearly localized L1 and the fluid L2. Our analysis reproduces successfully the coverage dependence of the activation gap, with a good quality fit to

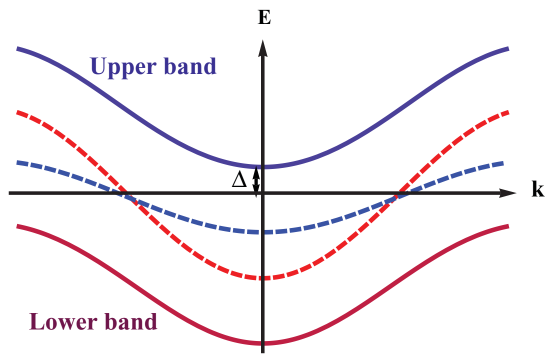

FIG. 1. (Color online) Sketch of the different dispersions for the hybridized bands (solid lines), the conduction electrons (upper dashed line), and the spinons (lower dashed line). The gap Δ is defined as the difference between the Fermi level and the bottom of the upper band.

the experimental data (see Figs. 3 and 4). The amplitude of the gap, however, departs from the observed experimental one by a factor of 10. Some discrepancy in the amplitude is to be expected, considering that the theory used an approximate Eliashberg scheme. Another analysis of this experiment has been carried out in Ref. 12. It is based on a cluster dynamical mean-field approach and leads to the identification of the transition observed experimentally with a band-selective Mott transition at which the upper band is gapped beyond the chemical potential. It accounts for the qualitative behavior of the effective mass and the energy scale T_0. However, the excitation gap as well as the absence of a critical regime in temperature are not discussed in this study.

As a second addition to our previous study, we make a simple *quantitative* prediction concerning the Weiss term θ in the magnetization behavior observed using NMR above T_0.[9] The latter is written

$$M(T > T_0) = \frac{C}{T-\theta}. \quad (1)$$

Because NMR provides a local measure of the nuclear magnetic susceptibility, we attribute the Weiss term to the Kondo coupling between the two layers. Within our model, and by definition of the Kondo breakdown QCP, these two layers decouple at the *real* QCP, at a coverage slightly smaller than for the QCP induced by the extrapolation of the quantum fluctuation regime. Accordingly, the Weiss term θ should vanish precisely at the coverage

$$N_I \approx 9.2 \text{ nm}^{-2} \quad (2)$$

where the real QCP is theoretically situated. This quantitative prediction is simple enough to be checked in future experiments on this system.

We start with the evaluation of the specific heat contribution. The mean-field theory considered in our study[10] consists of a two-band model with an effective hybridization $V_{\text{eff}} \equiv Vb$, where V is the bare hybridization and b is the expectation value for the holon operator, between the spinons and the conduction electrons. Diagonalization of the corresponding Hamiltonian results in two upper $(+)$ and lower $(-)$ hybridized bands with dispersions $E_{\mathbf{k}\pm}$.

As discussed in our previous study of this system,[10] the bare hybridization at the QCP is already very strong compared to other energies of the model. Further, at the QCP, the f band is half filled and the upper band is constrained to stay below the Fermi level. However, as soon as the effective hybridization sets in strongly, we enter very soon a regime where that band is empty and a gap opens. This happens in the hybridized phase before the QCP is reached, which is consistent with the experimental observation.[9] The gap is defined simply as

$$\begin{aligned}\Delta &\equiv E_+(-D)\\ &= \tfrac{1}{2}\big\{-(1+\alpha')D + \epsilon_f - \mu\\ &\quad + \sqrt{[-(1-\alpha')D - \epsilon_f - \mu]^2 + 4V_{\text{eff}}^2}\big\}. \quad (3)\end{aligned}$$

Here D is the half bandwidth of the conduction electrons, ϵ_f and μ are the chemical potentials of the f spinons and c electrons, respectively, and α' is the effective ratio between the bandwidths of the two fermionic species.

The gapped band contribution to the mean-field free energy reads

$$F_+ = -2T \sum_{\mathbf{k}, i\omega_n} \ln(-i\omega_n + E_{\mathbf{k}+}), \quad (4)$$

where $\beta = 1/T$.

The evaluation of (4) is straightforward. It is easily shown that at very low temperatures, such that $\Delta \gg T$, the specific heat contribution of the gapped band simplifies to

$$C_+ = \frac{\lambda}{T} e^{\frac{-\Delta}{T}}, \quad (5)$$

where

$$\lambda \equiv \frac{\rho_0}{\alpha'}\Delta^2 \left(1 + \alpha' + \frac{1-\alpha'}{\sqrt{4\alpha' V_{\text{eff}}^2}}[\epsilon_f - \alpha'\mu - (1-\alpha')\Delta]\right),$$

ρ_0 being the density of states at the Fermi level of the conduction electrons. The expression (5) is our final result. Notice that the functional form there is different from the one used in C_{expt} by the experimentalists. In particular, the coefficient of the exponential depends on temperature as $1/T$. In the following, we discuss our results. The theoretical gap is given by the expression (3). It turns out that it has the same coverage dependence, up to a constant factor of 10, as the one extracted experimentally from C_{expt}[9] (see Fig. 2).

As emphasized in the introduction, the values of the experimental gap depend strongly on the fitting function used by the experimentalists to capture the low-temperature behavior of the specific heat. Thus, what has to be done is to try fitting the gapped part of the specific heat using the expression (5) obtained within our model. We consider now the gapped part of the experimental data for specific heat, $C_g(T) = C_{\text{expt}}(T) - \gamma T$; we find its temperature dependence rather well reproduced by our analytical form (5), provided the values of the gap Δ are corrected by a constant factor of 10. Figures 3 and 4 show this perfect agreement for coverages $N = 8.25$ and 9.00 nm^{-2}.

The analytic form (5), deduced from our model, seems indeed to be adequate to describe the observed low-temperature behavior of the gapped specific heat. One can refine the

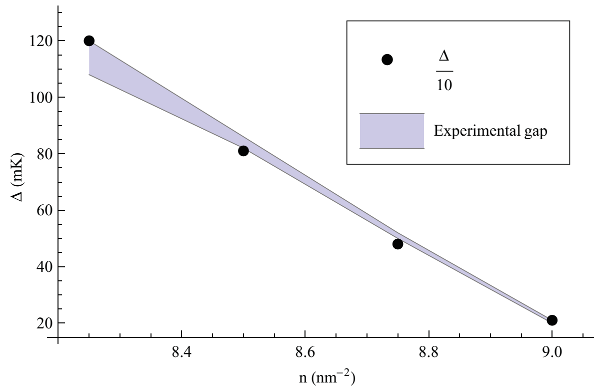

FIG. 2. (Color online) Comparison between Δ and the experimental gap Δ_{expt} (Ref. 9) for different doping levels.

effective magnitude of the gap Δ by assuming that it has nodes in the momentum space, i.e., it does not open uniformly along the Fermi surface. Indeed, the layer L1 solidifies into a commensurate lattice with respect to the underlying substrate, which has a triangular lattice.[9] Thus, the bare hybridization V could be momentum dependent, following one irreducible representation of the lattice symmetry group, whereas it is assumed to be local in our model.[10] Investigation in this direction and its possible implication have not been performed yet. In this paper as well as in a series of previous studies[10] we have used the KB theory of quantum criticality in order to study the phase transition in ^{3}He bilayers. One important feature of our model, generally poorly understood, is the key role of frustration around the phase transition. In the Kondo lattice model, there are two main mechanisms to quench entropy (see Fig. 5). One is through the formation of Kondo singlets, which after merging together form the heavy Fermi liquid, and the other one is through entanglement of the spins between themselves, resulting in the formation of the spin liquid.[13] The quenching of the entropy through spin-liquid formation is highly sensitive to the presence of frustration in the system, which is indeed present in ^{3}He bilayers due to ring exchange interactions.[14,15] We want to emphasize that in our KB model for ^{3}He bilayers, the QCP is driven by the spin liquid and

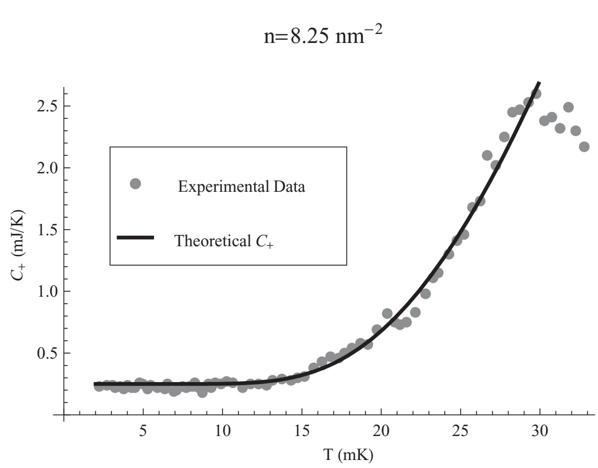

FIG. 3. Comparison between C_+ and the gapped part of the experimental data (Ref. 9) for $n = 8.25$ nm^{-2}.

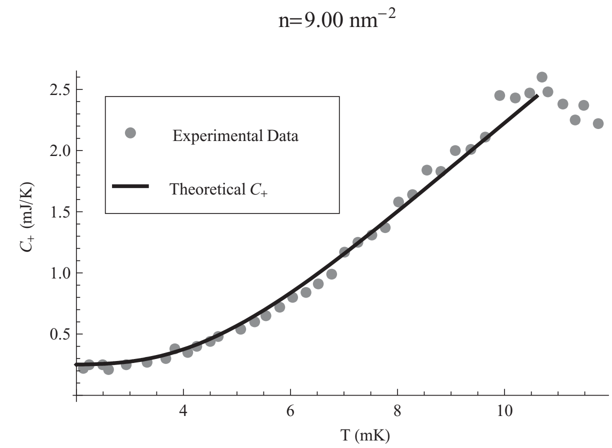

FIG. 4. Comparison between C_+ and the gapped part of the experimental data (Ref. 9) for $n = 8.25$ nm^{-2}.

not by the formation of the Kondo singlets. The entropy is quenched through the formation of a spin liquid while the phase transition toward the heavy Fermi liquid is associated with the breakdown of the Kondo effect.

This observation enables us to make a quantitative theoretical prediction. One might be able to extract the Kondo scale experimentally, for example through a nuclear magnetic resonance experiment where the relaxation of the first layer to the bath of free fermions made of the second layer is predominant. A Curie-Weiss law for the susceptibility is expected there, where the Curie-Weiss term θ can be interpreted as the Kondo scale of the system.

One of the results of our previous investigation of this model[10] is that two apparent QCPs are present. We identified the experimental one with the extrapolation of a finite-temperature regime of fluctuations. We claim, however, that the real QCP is occurring "before" the experimental one, when coverage is increased.

If our model is correct, the scale θ should go to zero at the "real" QCP, and not at the experimental one. Concretely this

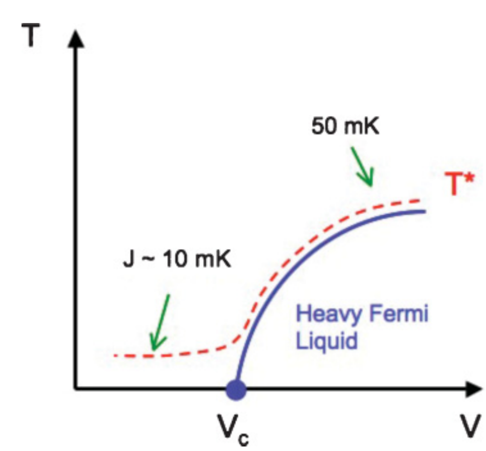

FIG. 5. (Color online) Phase diagram of the Kondo breakdown QCP. There are two successive quenchings of the entropy, one at T_0 due to the formation of the spin liquid and the other at the phase transition temperature. The Kondo scale is associated with the phase transition to the heavy Fermi liquid and should vanish at the QCP.

means that within our theory, θ vanishes for this experimental setup at the coverage

$$n_I \approx 9.2\ \mathrm{nm}^{-2}.$$

This is a strong prediction of our model and we hope that it can be tested experimentally in the very near future. The system studied, ^{3}He bilayers, in our series of papers, Ref. 10 and the present one, is one of the simplest physical ones, with negligible spin-orbit interaction and no crystal-field interactions. The beauty of the experiment in Ref. 9 is that it shows that such a system has qualitatively the same physics as HFCs and may serve thus as a test physical system for theories of quantum criticality in these complex systems.

In this Brief Report, we addressed one final feature of the experiment on this system, namely, the presence of an activation gap in the low-temperature behavior of the specific heat in the heavy-FL phase. We derived an analytic expression for the specific heat within our model, different from the one used by experimentalists. The behavior of the specific heat at low temperature is successfully reproduced, as well as the coverage dependence of the gap, albeit the latter has to be corrected by a constant factor of 10. This is expected within the Eliashberg treatment used here and may be a hint for a refinement of our model or a reevaluation of the parameters used in our previous study.

The KB model is successful and strong enough to account for many of the features of the experiment and make some quantitative predictions to be tested in future experiments on this system. It is now up to new experimental data to validate it or not and to different theoretical models to propose an alternative interpretation of the experiment.

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