Activation gap in the specific heat measurements for $^3$He bi-layers

A. Rançon-Schweiger$^1$, A. Benlagra$^2$ and C. Pépin$^{3,4}$

$^1$LPTMC, CNRS UMR 7600, Université Pierre et Marie Curie, 75252 Paris, France.
$^2$Institut für Theoretische Physik, Universität zu Köln, Zülpicher Str. 77, 50937 Köln, Germany.
$^3$Institut de Physique Théorique, CEA, IPht-CNRS,URA 2306, F-91191 Gif-sur-Yvette, France.
$^4$International Institute of Physics, Universidade Federal do Rio Grande do Norte, 59078-400 Natal-RN, Brazil

(Dated: February 18, 2011)

Recently, attention has been given to a system of $^3$He bi-layers where a quantum criticality similar to the one in heavy fermion compounds has been observed [Science 317, 1356 (2007)]. In our previous analysis [Phys. Rev. B 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 $\Delta$ 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.

PACS numbers: 71.27.+a, 72.15.Qm, 75.20.Hr, 75.30.Mb

I. INTRODUCTION

In recent years, the peculiar and non standard properties observed in heavy fermion compounds (HFC) 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. However, the difficulty with HFC 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 to rule in favor of a theoretical model or another.

Lately, a quantum criticality similar to the one in HFC has been observed in a much simpler system: $^3$He bilayers adsorbed on a composite substrate of graphite pre-plated with two solidified layers of $^4$He. Upon increasing the total coverage $N$ of $^3$He atoms, the system seems to undergo a quantum transition, at a critical coverage $N_{cr}$ ≈ 9.95 nm$^{-2}$, separating a regime where the two layers are hybridized and form a bi-fluid with heavy Fermi liquid (FL) properties to a regime where they are completely decoupled, the first one (L1) forming a frustrated 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 HFC 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 to use one of the models proposed to describe quantum criticality in HFC, namely the so-called Kondo breakdown (KB) scenario, to account for the remarkable features of the experiment. In that study, the $^3$He bi-layers 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 the ones of L2 with the itinerant conduction electrons. Remarkably, our study identifies the real QCP as occurring at a coverage of $N_f \approx 9.2$ 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 $N_f \approx 9.2$ 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 been also called 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 QCP’s 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. 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. The presence of an excitation gap is needed to fit the specific heat measurements in the hybridized phase according to \( C_{\text{exp}} = \gamma T + \gamma_2 e^{-\Delta_{\text{exp}}/T} \), where the first term is usual for a FL with \( \gamma \) proportional to the effective mass of the quasi-particles. The second term defines the experimental activation gap \( \Delta_{\text{exp}} \). Within our theory, the gap originates from an excitonic mechanism between the heavy and the light band of the system. The gap \( \Delta \) 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 the experimental data (see Figures 5 and 1). The amplitude of the gap, however, departs from the observed experimental one by a factor of ten. 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. [11].

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 a very simple 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

\[
\Delta \equiv E_+(-D) = \frac{1}{2} \left[ -(1 + \alpha')(D - \epsilon_f - \mu) \right] + \sqrt{\left[ -(1 - \alpha')D - \epsilon_f - \mu \right]^2 + 4\epsilon_{ff}^2}.
\]

Here \( D \) is the half bandwidth of the conduction electrons, \( \epsilon_f \) and \( \mu \) are the chemical potentials of the \( f \)-spinons and \( c \)-electrons respectively and \( \alpha' \) is the effective ratio between the bandwidths of the two fermionic species.

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

\[
F_+ = -2T \sum_{k,\omega_n} \ln(-i\omega_n + E_{k+}),
\]

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

\[
C_+ = \frac{\lambda}{T} e^{-\Delta/T},
\]

where

\[
\lambda \equiv \frac{\rho_0 \Delta^2}{\alpha'} \left[ 1 + \alpha' + \frac{1 - \alpha'}{\sqrt{4\alpha' V_{\text{eff}}}} (\epsilon_f - \alpha' \mu - (1 - \alpha') \Delta) \right].
\]
\( \rho_0 \) 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 for \( C_{\text{exp}} \) 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 \( (6) \). It turns out that it has the same coverage dependence, up to a constant factor of ten, of the one extracted experimentally from \( C_{\text{exp}} \) (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, \( C_g(T) = C_{\text{exp}}(T) - \gamma T \), we find its temperature dependence rather well reproduced by our analytical form \( (5) \), provided the values of the gap \( \Delta \) are corrected by a constant factor of ten. Figures 3 and 4 below, show this perfect agreement for coverages \( N = 8.25, 9.00 \text{ nm}^{-2} \).

The analytic form \( (5) \), deduced form our model, seems indeed to be adequate to describe the observed low temperature behavior of the gapped specific heat. One can refine the effective magnitude of the gap \( \Delta \) assuming that it has nodes in the momentum space, i.e. it doesn’t 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 bi-layers. 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 are forming the heavy Fermi liquid, and the other one is through entanglement of the spins between themselves, resulting in the formation of the spin liquid\(^12\). 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 bi-layers due to ring exchange interactions\(^13,14\). We want to emphasize that in our KB model for \(^3\)He bi-layers, the QCP is driven by the spin liquid and not by the formation of the Kondo singlets. The entropy is quenched through the formation of a spin liquid while the phase transition towards 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 $\theta$ can be interpreted as the Kondo scale of the system.

One of the results of our previous investigation of this model 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 $\theta$ should go to zero at the “real” QCP, and not at the experimental one. Concretely it means that within our theory, $\theta$ vanishes for this experimental set up at the coverage

$$n_f \approx 9.2 nm^{-2}.$$  

It is a strong prediction of our model and we hope that it can be tested experimentally in a very near future. The system studied, $^3$He bi-layers, 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 HFC and may serve thus as a test physical system for theories of quantum criticality in these complex systems.

In this manuscript, we addressed one final feature of the experiment on this system, namely the presence of an activation gap in the low temperatures behavior of the specific heat in the HFL 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 ten. This is expected within the Eliashberg treatment used here and may be a hint for a refinement of our model or a re-evaluation of the parameters used in our previous study.

The KB model is successful and strong enough to account for main 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.

**Acknowledgments**

This work was supported by the French National Grant ANR26ECCEZZZ. We acknowledge fruitful discussions with J. Saunders and I. Paul.

1. G. Stewart, Rev. Mod. Phys. 56, 755 (1984); 73, 797 (2001).
2. H. v. Löhneysen et al. Rev. Mod. Phys. 79, 1015 (2007).
3. P. Coleman et al., J. Phys. Cond. Matter 13 R723 (2001).
4. T. Moriya and J. Kawabata, J. Phys. Soc. Jpn. 34, 639 (1973); T. Moriya and J. Kawabata, J. Phys. Soc. Jpn. 35, 669 (1973); J. A. Hertz, Phys. Rev. B 14, 1165 (1976); A. J. Millis, Phys. Rev. B 48, 7183 (1993).
5. Q. Si, S. Rabello, K. Ingersent and J. L. Smith, Nature 413, 804 (2001); ibid. Phys. Rev. B 68, 115103 (2003)
6. T. Senthil, M. Vojta, and S. Sachdev, Phys. Rev. B 69, 035111 (2004).
7. I. Paul, C. Pépin and M.R. Norman, Phys. Rev. Lett. 98, 026402 (2007), ibid. Phys Rev B, 78, 035109 (2008).
8. C. Pépin, Phys. Rev. Lett. 98, 206401 (2007), C. Pépin, Phys. Rev. B, 77, 245129 (2008).
9. M. Neumann et al., Science 317, 1356 (2007).
10. A. Benlagra and C. Pépin, Phys. Rev. Lett. 100, 176401 (2008), ibid. Phys. Rev. B 79, 045112 (2009).
11. K. S. D. Beach and F. F. Assaad, arXiv:0905.1127v1
12. At lower temperature of course, the occurrence of phase transitions, of magnetic type for example, can quench the entropy further.
13. M. Roger, Phys. Rev. Lett. 64, 297 (1990).
14. G. Misguich, P. Bernu, C. Lhuillier and D. Waldtmann, Phys. Rev. Lett 81, 1098 (1998).
15. J. Boronat, J. Casulleras, V. Grau, E. Krotscheck and J. Springer, Phys. Rev. Lett. 91, 085302 (2003).