Research Article

Spin vs charge excitations in heavy-fermion compounds♠

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It is pointed out that the answer on the question about the role played by spin and charge excitations will help to solve the physical origin of the heavy-fermion phenomena. Our answer is that neutral spin-like excitations are responsible for the heavy-fermion phenomena whereas the role of the charge excitations is negligible.

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I. INTRODUCTION

The microscopic origin of the heavy-fermion phenomena and the nature of quasiparticles, despite of 25 years of very intensive theoretical and experimental studies, is still a subject of controversy [1–9]. The aim of this paper is to point out that neutral spin-like excitations are responsible for the heavy-fermion phenomena whereas the role of charge excitations is negligible (An extra remark - this sentence has been underlined by the Chairman of SCES-02 in the rejected copy with a note -WRONG!!).

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II. THEORETICAL UNDERSTANDING

Characteristic heavy-fermion (h-f) phenomena like a large low-temperature specific heat, a non-magnetic state (we would rather say a weakly-magnetic state) anticipated from a Pauli-like low-temperature susceptibility and the anomalous resistivity have been basically discussed in terms of the Fermi liquid (FL). In the FL picture the excitations are charged and they become possible due to the strong hybridization of f-electron and conduction-electron states. In the FL picture the f states lie at the Fermi level and, as a consequence of the strong hybridization, the number of the f electrons, n_f, becomes not integer. The occupation number n_f is the important factor in the FL description as many physical properties are renormalized by the term $(1\text{-}n_f)^{-1}$. Kondo temperature is directly related to this factor as one can read from Eq. 15 in Ref.[3], p.608. In this picture the disappearance of the local moment is related to the unoccupied f states [3]. Thus, in the FL picture a deviation of the occupation number n_f from the integer value and the charge excitations play the fundamental role in formation of heavy-fermion phenomena.

In our understanding of heavy-fermion phenomena we think that even in Ce, U, Yb compounds, exhibiting the h-f phenomena, the f states lie much below E_F like in conventional rare-earth compounds [10]. A large low-temperature specific heat is related to the magnetic excitations well understood in conventional rare-earth compounds in case of the Kramers electron systems, i.e. systems with an odd number of the f electrons. Such the odd number electron system is realized in case of the $f^1(Ce^{3+})$, f^3 (U^{3+}), f^{13} (Yb^{3+}) systems. All of them have the Kramers doublet charge-formed (CF) ground state. This double degeneracy has to be removed before the system reaches 0 K and, according to us, the h-f compounds are compounds with extremely small magnetic temperatures. The removal of the Kramers degeneracy is equivalent to the time-reversal symmetry breaking (on the atomic scale) and to the formation of the magnetic state (on the atomic scale). These phenomena can be well discussed within the Quantum Atomistic Solid-State Theory (QUASST), that points out that f atoms preserve much of their atomic properties becoming the full part of a solid [11]. In our model there is no single Sommerfeld coefficient γ within the wide, but still below, say, 2 K, temperature range. In contrary to a very loosely term "non-Fermi liquid behaviour" QUASST predicts that the ground state of the h-f compound is magnetic, i.e. with the broken time-reversal symmetry. Moreover, it is the state with a low local symmetry and the broken translational

symmetry, in a sophisticated manner, leading to the differentiation of the f atoms with respect to the shape of the CF ground state and of the magnetism. It causes that the magnetic state is not coherent with respect to temperature, space and the local moment direction, in the sense that local magnetic states, marked by the splitting of single-ion Kramers doublet, appear at slightly different temperatures at different sites and with different direction of the local moment. According to this understanding the excitations are neutral, spin-like and of very small energy. They mimics a spin-liquid with non-trivial properties like the strong spin-lattice coupling and the substantial orbital contribution to the magnetic moment. In the theoretical description it will appear as the need for the attributing the spin with unusual highly anisotropic properties. This fact causes that the local excitations, with the reversal of the local moment, is not longer a local event. The Sommerfeld coefficient γ can be extremely large at low temperatures - Ropka has calculated γ of 25 J/K mol [12]. The lower magnetic temperature, i.e. the temperature where the splitting of the Kramers doublet appears, the larger γ can be. It is very important that our model overcomes the Nozieres exhaustion argument [13] about insufficient number of conduction electrons to compensate all localized moments, of value comparable to that observed in the paramagnetic state, by means of the spin-compensation mechanism. We have shown that the crystal-field (charge) mechanism, related to the anisotropic charge distribution at the vicinity of the paramagnetic cation, is much more effective than the spin-compensation mechanism [14] in the reduction of the local magnetic moments, even down to almost zero value.Anisotropic charge distribution around the $4f$-cation can produce the crystal-electric-field (CEF) ground state with a quite small magnetic moment even in case of the Kramers system.

III. DISCUSSION

We take the growing evidence for the Non-Fermi -Liquid behaviour revealed in rapidly growing number of compounds as the confirmation of our understanding of the heavy-fermion phenomena. Our understanding, with the integer number of f electrons, concurs with theoretical results of Doradzinski and Spalek who came to a number of f electrons so close to 1 as 0.995 [9]. It is worth to remember that values for n_f were given in years 1985-1990 as about 0.70-0.80, see [7], for instance - thus a value of 0.995 we take as practically 1. Within our model we have managed to describe low-temperature specific heat of $Nd_{2-x}Ce_xCuO_4$ as originating from excitations to the conjugate Kramers state of the

Nd^{3+} ions [15] despite that $Nd_{2-x}Ce_xCuO_4$ has been announced in 1993 as a new class of heavy-fermion superconducting compounds. We have managed to describe an anomalous temperature dependence of the quadrupolar splitting in $YbCu_2Si_2$ as the conventional crystal-field effect on the Yb^{3+} ions [16]. It was an important result as this anomalous dependence was given as the conclusive evidence for the hybridization of f electrons and conduction electrons. Thus, the good description within the crystal-field model has abolished the hybridization mechanism and that a n_f value of 0.82 (hole) as completely artificial because the Yb ion behaves in $YbCu_2Si_2$ as the $4f^{13}$ system. The existence of 3 f electrons as highly-correlated $5f^3$ system has been proved in heavy-fermion superconductor UPd_2Al_3 by observation of well defined CEF-like states [17].

QUASST can be applied not only to intermetallics, where the h–f behaviour was found originally, but also to insulating rare-earth systems, known as low-carrier systems (Sm_3Se_4, Yb_3S_4) and to nuclear systems (^{3}He). Our model, developed already in 1994, has predicted the possibility of h-f phenomena in 3d and 4d compounds - the h-f behaviour has been discovered in LiV_2O_4 in 1997, indeed. QUASST predicts smooth crossover from the heavy-fermion state to the conventional localized-moment state with the Curie-Weiss law fulfilled, in agreement with observations. The f electrons, being localized in an integer number n depending on the partners and on the composition of a considered compound, take the active role in the solid-state bonding, via the conventional coulombic interactions.

Finally, we are at the Conference devoted to Strongly-Correlated Electron Systems. In our atomic-like approach the correlations among the f electrons are taken to be really very strong - thanks them, atomic-like terms and multiplets as well as three Hund's rules are fulfilled.

IV. CONCLUSIONS

We claim that heavy-fermion phenomena are caused by neutral spin-like excitations, whereas the role of the charge excitations is negligible. The ground state of heavy-fermion compounds is magnetic with the time-reversal symmetry broken at the atomic scale.

An extra note added March 17, 2003. We take a recent paper of Zwicknagl, Yaresko and Fulde, Phys. Rev. B 65 (2002) 081103, that came to our attention during the SCES-02 Conference, with a novel model for heavy-fermion phenomena with two localized f electrons as

confirmation of our 10-year understanding of heavy-fermion phenomena with the importance of crystal-field states. In particular, in the situation of Prof. P. Fulde who by twenty years advocated for the itinerant f-electron origin of heavy-fermion phenomena. It is worth to remind that in the SCES-94 Conference the crystal-field theory has been rejected from the conference presentation as not at all related to the heavy-fermion physics. All members of the International Advisory Committee have been informed about this abnormal situation but noone of them react in order to fulfill normal scientific rules within the magnetic community. Moreover, our CEF approach to heavy-fermion phenomena has been continuously rejected by Fulde, acting as the Editor, from publication in Zeitsch. Physik B (e.g. No MS606/94). Thus, we welcome this recent Zwicknagl *et al.* paper admitting the existence of CEF states in heavy-fermion and uranium compounds, though we think that the treatment of CEF in this paper is very oversimplified. There still is a problem of the existence of the $5f^2$ or $5f^3$ system in UPd_2Al_3. In contrary to the $5f^2$ system considering by Zwicknagl et al. there is large evidence for the $5f^3$ system. This evidence was published already in 1992, see references in Ref. [17], but Zwicknagl *et al.* have ignored it.

♠ This paper has been submitted 31.05.2002 to Strongly Correlated Electron Conference in Krakow, SCES-02 getting a code NFL023. It has been presented at the Conference, but it has been rejected by the Chairman of SCES-02.

The paper has been given under the law and scientific protection of the Rector of the Jagellonian University in Krakow, of the University of Mining and Metallurgy and of the Polish Academy of Sciences.

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