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CEF states in heavy-fermion superconductor UPd_2Al_3 ☆

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Abstract

The CEF-like excitations in UPd_2Al_3 revealed in neutron-scattering studies have been found consistent with earlier prediction on basis of the overall temperature dependence of the specific heat. The specific heat analysis yields 5 doublet states at energies 0, 100, 150, 350 and 600 K associated with the U^{3+} configuration. In fact, these states are similar to those observed in isostructural $NdNi_5$ though the overall CEF splitting is more than two times bigger. The observation of the localized states by such different techniques and their consistency provides strong argument for the applicability of the crystal-field (CEF) theory to (some) actinide compounds like it was found for UGa_2 and $NpGa_2$.

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The discovery in 1976 of heavy-fermion (h-f) phenomena associated with the extraordinarily large specific heat at low temperatures ($CeAl_3$) has challenged the magnetic community for their origin. The scientific intriguity has increased in 1991 when the h-f phenomena has been found in UPd_2Al_3 [1] because of the coexistence of the h-f phenomena and the large magnetic moment of about $1.5\mu_B$ ($T_N = 14$ K). Moreover, UPd_2Al_3 exhibits superconductivity below 2 K. Owing to these properties UPd_2Al_3 is really unique. Despite of very popular theories of h-f phenomena related with the delocalization of f electrons [2,3] there were approaches [4,5] offering the understanding of UPd_2Al_3 with the crystal-field (CEF) approach i.e. with keeping integer value of f electrons; 2 for the U^{4+} ion [4] or 3 for the U^{3+} ion [5].

The aim of this paper is to point out that the existence of the localized f electrons revealed by inelastic-neutron-scattering experiments [6] in the heavy-fermion UPd_2Al_3 is consistent with the earlier prediction on basis of the overall temperature dependence of the specific heat with the U^{3+} configuration.

The search for the crystal-field excitations in UPd_2Al_3 has been undertaken by the group of Steglich [6] with the purpose to verify the U^{4+} states proposed in Ref. [4]. The inelastic-neutron-scattering experiments have revealed at 25 K the existence of the crystal-field excitations with energies of 7 and 23.4 meV. This experiment at 150 K has revealed further excitations at 3 and 14 meV at the energy-loss side and at 7 meV at the energy-gain side.

The observation of CEF excitations as well as their energies are in good agreement with those predicted on basis of the specific-heat analysis [5]. This analysis has yielded the energy levels at 0, 100, 150, 350 and 600 K, i.e. at 0, 8.6, 12.9, 30.1 and 51.7 meV (Fig. 1). All of them are Kramers doublet owing to the assumed U^{3+} configuration. According to the derived energy-level scheme the observed transitions at 25 K are the transitions from the ground state to the first and to the third excited state as is shown by arrows in Fig. 1. The transitions detected at 150 K correspond to the transition from the first excited state to the second and to the third excited state as then the first excited state is already substantially thermally

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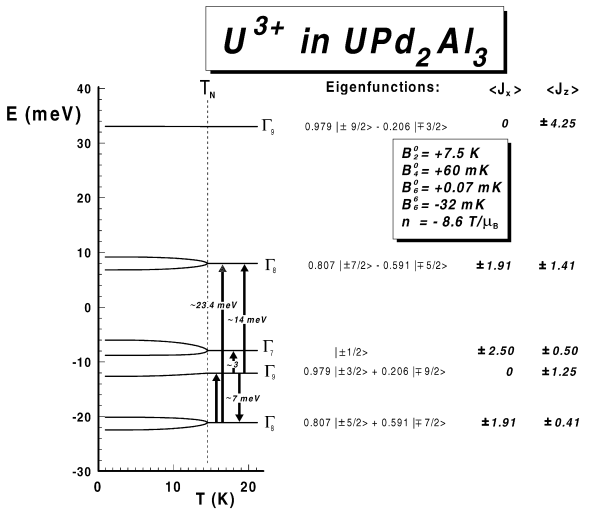

Fig. 1. Energy level scheme of the U^{3+} ion in UPd_2Al_3. The arrows indicate transitions that we have attributed to the excitations revealed by inelastic-neutron-scattering experiments [6].

populated. The transition at 7 meV at the energy-gain side corresponds to the transition from the first excited state to the ground state.

For the description of electronic and magnetic properties we have applied a single-ion-like Hamiltonian:

$$H = H_{CF} + H_{f-f} = \sum\sum B_n^m O_n^m + ng^2\mu_B^2 \times\left(- J\langle J\rangle + \frac{1}{2}\langle J\rangle^2\right).$$

The first term is the crystal-field Hamiltonian for the hexagonal symmetry with $B_2^0 = +7.5$ K, $B_4^0 = +60$ mK, $B_6^0 = +0.07$ mK and $B_6^6 = -32$ mK [5]. The second term takes into account the intersite spin-dependent interactions that produce e.g. the magnetic order below T_N what is seen in Fig. 1 as the appearance of the splitting of the Kamers doublet.

Such good agreement with predictions derived for the U^{3+} ion in UPd_2Al_3 on basis of completely different experiments indicate the large physical adequancy of the derived fine energy level scheme and of the U^{3+} state. In the situation of the fundamental scientific controversy about the localization/delocalization mechanism for the formation of the h-f state the successful result of the search for the crystal-field excitations of Krimmel et al. is of great importance. This fact we are taking as strong support for the validity of the crystal-field model for the h-f phenomena [7]. According to us the crystal-field model gives the microscopic base for the spin-fluctuation mechanism of the heavy-fermion phenomena, that is in fundamental opposition to the charge-fluctuation mechanism [2,3]. In this model the large specific heat results from low-energy excitations to the conjugate Kramers state whereas the h-f particles are neutral spin-like excitations. This model works for systems containing the f^1 (Ce^{3+}), f^{13} (Yb^{3+}), f^3 (Nd^{3+} and U^{3+} ions) and other odd number electronic systems, both in ionic and intermetallic compounds. Obviously, the finding of the CEF excitations in the heavy-fermion UPd_2Al_3, largely ignored in nowadays in-fashion solid-state theories, calls for a more advanced solid-state physics theory.

We would like to point out that the existence of the localized f electrons does not contradict the formation of the cooperative phenomena like magnetism or superconductivity. In system like UPd_2Al_3 the metallic properties are assured by conduction electrons made by among others three outer uranium electrons. Within the individualized-electron model in intermetallics there coexists a few electronic subsystems, conduction and f-electron system, that strongly interacts but without charge exchange between them. In this picture f atoms in a solid preserve much of their individual atomic properties. Then the U^{3+} ion has the fine electronic structure similar to that observed in Ni^{3+}-ion compounds [8]. In fact, the structure found in UPd_2Al_3 is very similar to that found for $NdNi_5$. Both compounds are hexagonal with the magnetic moment directed perpendicular to the hexagonal c-axis. These phenomena are accounted for by the crystal field, that in UPd_2Al_3 is more than two times bigger than in $NdNi_5$ (the overall splitting amounts to 54 and 21 meV, respectively). The CEF parameters for $NdNi_5$ of $B_2^0 = +3.35$ K, $B_4^0 = +14.5$ mK, $B_6^0 = -0.35$ mK and $B_6^6 = -13.5$ mK are quite similar to that of UPd_2Al_3. B_2^0, that determines the overall CEF splitting is more than two times smaller.

In conclusion, it is pointed out that the experimentally observed crystal-field excitations in the heavy-fermion UPd_2Al_3 are consistent with the U^{3+} state. It makes that UPd_2Al_3 is the second, apart UPd_3, uranium compound in which CEF states have been unambiguously revealed. The existence of the localized f states in heavy-fermion superconductor UPd_2Al_3 is taken as a strong support for the validity of the crystal-field model for the h-f phenomena in which the crystal-field realization of the Kramers-doublet ground state is the basic ingredient. Very similar way of thinking has been presented by Furrer and his coworkers for the heavy-fermion superconductor $Nd_{2-y}Ce_yCuO_4$ [9]. The observation of the localized states by such different techniques and their consistency provides strong argument for the applicability of the CEF theory to (some) actinide compounds like it was found for UGa_2 and $NpGa_2$ [10].

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