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Kramers systems with weak spin-dependent interactions

R.J. Radwański *,

Inst. Physics & Informatics, Pedagogical University, 30-084 Kraków, Poland
Center for Sodid State Physics, Św. Filip 5, 31-150 Kraków, Poland

Abstract

Magnetic properties of a paramagnetic Kramers f^3 subsystem under charge interactions of the hexagonal symmetry have been examined in the combination with weak spin-dependent (S-D) interactions for the case of the weakly-magnetic charge-formed ground state. The Kramers systems with weak S-D interactions exhibit particular phenomena like large specific heat at low temperatures.

In Kramers systems (systems with an odd number of electrons or, more general, of fermions) the charge-formed (CF) ground state is a Kramers doublet. The Kramers doublet states are split in the magnetically ordered state. The observed magnetic-ordering temperature T_o is usually higher than, say, 3 K. For example, for $ErNi_5$ and $NdNi_5$ intermetallics, containing Kramers system f^{11} (Er^{3+}) and f^3 (Nd^{3+}), T_c amounts to 9.1 [1] and 7.2 K [2], respectively. Their magnetic and electronic properties, including the value of T_c and the ordered moment, are well reproduced by crystalline-electric field (CEF) and molecular-field (MF) calculations, i.e., within the localized f-electron picture. The spontaneous moment in $NdNi_5$ amounts to 2.1 μ_B/f.u., i.e., to 60% of that expected for the Nd^{3+} free ion. This reduction has been fully accounted for as an effect of higher-order CEF interactions [3]. However, it has been believed that, within the localized picture, the *small value* for the ordering temperature and the magnetic moment cannot be realized *for Kramers systems*. Recent studies of the bound states of the paramagnetic Kramers ion under complex CEF interactions have proved that charge interactions of the hexagonal symmetry can fully suppress the local moment even in the case of the Kramers system [3] revealing a strong correlation between the magnetic state of the ground state of the f^n localized subsystem and the (anisotropic) charge distribution at the vicinity of the f shell. It means that any value for the local moment can be realized as the effect of electric charge interactions.

The aim of this paper is to study the removal of the Kramers degeneracy of the f^3 system in case of weak spin-dependent (S-D) interactions. The f^3 system (3 f electrons) is relevant for the Pr^{2+}, Nd^{3+}, U^{3+} or Np^{4+} ions.

* Corresponding author. Fax: +48-12-336146.

The f^3 system within the Russel–Saunders scheme is characterized by the total angular momentum J of 9/2 and the ground multiplet $^4I_{9/2}$. Charge interactions of the hexagonal symmetry lift the 10-fold degeneracy and produce five Kramers doublets [3]. This Kramers degeneracy, being associated with time-reversal symmetry, cannot be removed by charge interactions. It can be only removed by an external or internal magnetic field, the latter resulting from S-D interactions [3]. The effect of charge and S-D interactions can be quantatively traced when they are approximated by the CEF approach and by the MF approach, respectively. The single-ion Hamiltonian of the R^{3+} ion is written [1,2]:

$$H_R = \sum \sum B_n^m O_n^m + ng^2\mu_B^2\left(-J\langle J\rangle + \tfrac{1}{2}\langle J\rangle^2\right).$$

It allows for calculations of bound/localized states of the f^n system, i.e., their positions and eigenfunctions.

A set of CEF parameters: $B_2^0 = +4.5$ K, $B_4^0 = -123$ mK, $B_6^0 = -2$ mK, $B_6^6 = -52.3$ mK and $n = 8.5\ T/\mu_B$ has been used in order to get values of T_o of 1 K and the ordered moment of $0.44\mu_B$. The ground state is $\Gamma_9 := -0.58|\pm 9/2\rangle + 0.81|\mp 3/2\rangle$ and the excited states are Γ_7 (82.4 K), Γ_8 (201 K), Γ_9 (374.9 K) and Γ_8 (850.7 K). The six-order term B_6^6 produces the hybridization of the atomic-like $|J,J_z\rangle$ states leading to the ground state with largely suppressed magnetic characteristics ($m = 0.38\mu_B$). Fig. 1 shows the formation of the singlet ground state of the f subsystem due to the combined action of the charge and S-D interactions. An energy gap appears at the magnetic ordering temperature reaching a value of 1.95 K at the absolute-zero temperature. This splitting is a source of a lot of excitations observed in specific-heat experiments, at temperatures comparable to the energy splitting, below 1 K in the present case.

Fig. 2 illustrates the formation of the local moment that is somehow typical for Kramers systems. The moments

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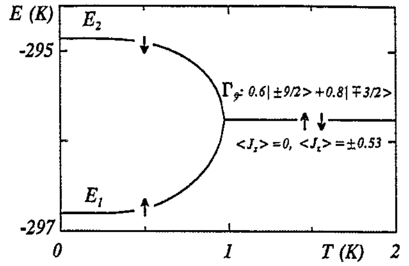

Fig. 1. The splitting of the CF ground state Γ_9 of the f^3 Kramers system by weak S-D interactions below T_0 of 1.0 K simulated by a set of CEF parameters of the hexagonal symmetry (see text). The ground state has weak magnetic characteristics with expectation values of $\langle J_z \rangle = \pm 0.53$ and $\langle J_x \rangle = 0$. Arrows indicate the direction of the magnetic moment associated with each local state.

associated with the CF Kramers twin-states have, in general, the opposite directions though with slightly different values. The excitation to the Kramers twin-state, due to the temperature, causes the reversal of the local moment. If conduction electrons are present in the solid, the reversal of the local moment implies, due to a Kondo-type interaction, the reversal of the oscillatory spin polarization of the conduction electrons leading to an effective binding of localized and conduction electrons. The thermally-induced local excitation is not any longer a local event. In comparison, the situation with strong S-D interactions without reversal of the moment is shown in Fig. 3 for $NdNi_5$. $NdNi_5$ is a conventional ferromagnet with T_c of 7.2 K pointing the strong S-D interactions that produce an internal field of 7.0 T [2]. As strong S-D interactions destroy the moment-reversal symmetry one comes to a rather striking conclusion that binding of localized and conduction electrons is much more effective in case of weak S-D interactions.

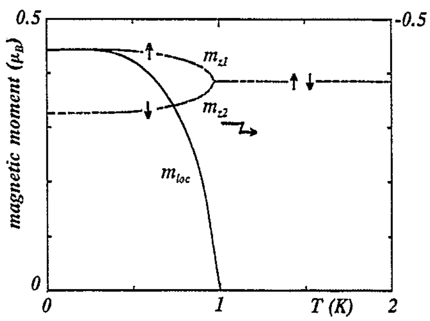

Fig. 2. Temperature dependence of the local magnetic moment (m_{loc}) and the moments associated with two Kramers twin-levels m_{z1} and m_{z2} in Fig. 1. Magnetic moments associated with two Kramers twin-states have the opposite sign (note that for the moment m_{z2} the left scale is relevant) that cancel exactly in the paramagnetic state. For weak S-D interactions moments become different remaining opposite directions. In the ordered state the value of the local moment increases due to the increasing difference in the population of the two Kramers twin-states (mainly) and increasing difference of their values.

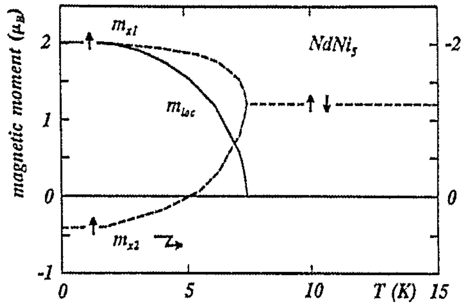

Fig. 3. Temperature dependence of the magnetic moment (as Fig. 2) for $NdNi_5$, a conventional ferromagnet with T_c of 7.2 K, with parameters $B_2^0 = +3.35$ K, $B_4^0 = +14.5$ mK, $B_6^0 = -0.35$ mK, $B_6^6 = -13.5$ mk and $n = 3.5\ T/\mu_B$. Below 5 K there is no reversal of the moment as the moments associated with two Kramers twin-states have the same direction.

Some important features concerning the Kramers systems could be summarized as follows: (i) the Kramers degeneracy has to be lifted before reaching the absolute-zero temperature; otherwise the 3rd thermodynamic law will be violated; (ii) the removal of the Kramers degeneracy is equivalent to the appearance of the local magnetic moment and of the internal magnetic field, (iii) the ground state of the Kramers system is always magnetic, though the associated local moment can be very small, (iv) in case of weak S-D interactions the system has difficulties in the removal of the Kramers degeneracy resulting in interesting phenomena at low temperatures.

In summary, some magnetic properties of a paramagnetic Kramers f^3 subsystem under charge interactions of the hexagonal symmetry and weak S-D interactions have been examined for the case of the weakly-magnetic CF ground state. An f^3 system with the local moment and the ordering temperature as small as $0.38\mu_B$ and 1 K, respectively, has been súccessfully simulated in the localized-electron approach. It has been pointed out that thermal excitations within the slightly-split Kramers doublet are associated with the reversal of the local magnetic moment that subsequently implies the reversal of the oscillatory spin polarization of conduction electrons. The binding mechanism of localized and conduction electrons is found to be more effective in case of weak S-D interactions, i.e., when S-D interactions do not change CEF levels. The revealed properties seem to be applicable to some f intermetallics exhibiting the heavy-fermion behaviour and to ^3He, that can be considered as a nuclear Kramers system.

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