MAGNETIC PHASE TRANSITIONS AND ORBITAL MAGNETISM IN THE MOTT INSULATORS $FeBr_2$, UO_2 AND IN $ErNi_5$*

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Magnetic phase transitions in $FeBr_2$, $ErNi_5$ and UO_2 have been found to be well described within the same quasi-atomic crystal-field approach with mean-field approximation for the exchange. The $\lambda-$type peak observed at the magnetic ordering temperature in the heat capacity is related to the time-reversal symmetry breaking at the atomic scale. The good description proves the existence of discrete atomic-like states, on the meV energy scale, both in $3d$, $4f$ and $5f$ systems confirming *a posteriori* the basic assumption of our theory. Our studies indicate that the intraatomic spin-orbit coupling and the orbital magnetism are indispensable for a physically adequate description of both electronic and magnetic properties.

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1. Introduction

In this paper we are interested in the magnetic phase transition in compounds containing atoms of transition metals of the iron group, of lanthanides and of actinides. These atoms contain $3d$, $4f$ and $5f$ open shells, respectively. The aim of this paper is to provide the evidence for the magnetic transitions involving the discrete quasi-atomic electron states in solid-state compounds containing open-shell atoms. We concentrate on the magnetic transition in $FeBr_2$, UO_2 and $ErNi_5$. For description of the magnetic transitions in all of these compounds we have applied the crystal field approach [1,2].

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2. Theoretical outline and results

According to the crystal-field approach the n electrons in the incomplete $3d/4f/5f$ shell form the highly-correlated intraatomic $3d^n$ /$4f^n$ /$5f^n$ electron system also when the paramagnetic atom becomes a part of a solid. These strong correlations among the incomplete-shell electrons are accounted for in zero-order approximation by two Hund's rules, that yield the ^5D ground term the $3d^6$ system in the Fe^{2+} ion in $FeBr_2$, the $^3H_4$ ground multiplet for the $5f^2$ system in the U^{4+} ion in UO_2 and the $^4I_{15/2}$ for the $4f^{11}$ system in the Er^{3+} ion in $ErNi_5$. Our interest in $FeBr_2$ is related to the fact that it is one of the strongest metamagnet. At 4.2 K an external field of 3.15 T causes a jump of the magnetization from the almost zero value to a very big value of 4.4 μ_B. This value exceeds the theoretical value of 4.0 μ_B, expected for the spin-only moment. $FeBr_2$ exhibits an antiferromagnetic (AF) ordering below T_N of 14.2 K. The appearance of the AF ordering is marked in the specific heat by a pronounced λ-type behavior. The magnetic phase transition in $FeBr_2$ has been found to be well described within the same atomic-like approach as has been successfully used for $ErNi_5$ [3]. However, we have to take into account a fact, that the spin-orbit coupling is much weaker in case of the $3d$ ions. The used Hamiltonian for the Hund's rules ground term (S=2, L=2) takes a form [4]:

$$\begin{aligned} H &= H_d + H_{d-d} \\ &= \sum\sum B_n^m \hat{O}_n^m + \lambda L \cdot S + n\left(-m\langle m\rangle + \tfrac{1}{2}\langle m\rangle^2\right) - m \cdot B_{\text{ext}}\,, \end{aligned} \quad (1)$$

where $m = -(L+2S)\mu_B$ is the magnetic-moment operator of the d-atom and n is the molecular-field coefficient. The first term is the full-symmetry CEF Hamiltonian. The magnetically-ordered state is calculated self-consistently. The optimal parameters found for $FeBr_2$ are: the octahedral CEF parameter B_4=+200 K, the spin-orbit coupling $\lambda = -150$ K, $n = -0.8K/\mu_B^2$ and the slight trigonal off-cubic distortion $B_2^0 = -30$ K. The calculated temperature dependence of the ordered magnetic moment and of the heat capacity are shown in Fig. 1 together with the temperature dependence of the energy of the three lowest localized states.

UO_2 is antiferromagnetic below $T_N = 30$ K. We have attributed the magnetism of UO_2 to the U^{4+} ions. The U^{4+} ion has 2 electrons forming the atomic-like system $5f^2$. According to Hund rules it is described by the quantum numbers L=5, S=1, and J=4. The used single-ion-like Hamiltonian takes the form:

$$\begin{aligned} H &= H_f + H_{f-f} \\ &= \sum\sum B_n^m \hat{O}_n^m + n g_L^2 \mu_B^2 \left(-J\langle J\rangle + \tfrac{1}{2}\langle J\rangle^2\right) + g_L \mu_B J \cdot B_{\text{ext}}\,, \end{aligned} \quad (2)$$

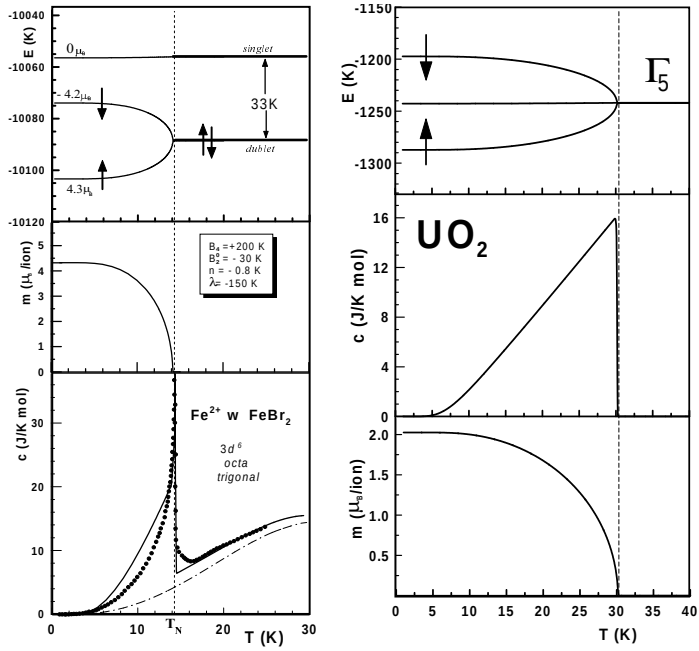

Fig. 1. Magnetic phase transition in $FeBr_2$. (a) temperature dependence of the 3 lowest energy levels; (b) temperature dependence of the magnetic moment of the Fe^{2+} ion; (c) temperature variation of the contribution of the d subsystem to the molar heat capacity. The points represent the experimental data [5].

Fig. 2. Magnetic phase transition in UO_2. (a) temperature dependence of the 3 lowest energy levels of the ground triplet Γ_5; (b) temperature variation of the f-subsystem contribution to the heat capacity of UO_2; (c) temperature dependence of the atomic magnetic moment of the U^{4+} ion.

where g_L is the Lande factor and the magnetic moment of the U ion is expressed as m=-$g_L J\mu_B$. The CEF parameters $B_4 = +733$ mK and $B_6 = +3.88$ mK, resulting from the cubic oxygen surroundings, yields the triplet Γ_5 as the ground state. These parameters provide the states in agreement with the inelastic-neutron-scattering data of Ref. [6]. The critical analysis of the CEF interactions in UO_2 performed in Ref. [7] shows that evaluation of CEF parameters, for description of the electronic structure on the energy scale of 160 meV was quite troublesome for more than 40 years. All of these results agree about the triplet Γ_5 (T_2) as the ground state. In Fig. 2 we present the calculated temperature dependence of the ordered magnetic moment and of the heat capacity, together with the temperature dependence of the three lowest energy levels composing the Γ_5 triplet. The value n of $-11.0K/\mu_B^2$ yields $T_N = 30$ K and the molecular field of 34 T at 0 K.

3. Conclusions

We have presented a microscopic description of the magnetic transition for two different systems containing partially filled $3d$ /$5f$ shell atom, namely $FeBr_2$ and UO_2 performed within the same atomistic concept as we have used before, Ref. [3], for rare-earth compound $ErNi_5$. In all of these compounds exist discrete low-energy states originating from the atomic-like highly-correlated electron system $3d^n$, $4f^n$ and $5f^n$. For the description of these states we took into account the orbital quantum number and the spin-orbit coupling. The λ-type peak observed at the magnetic ordering temperature is related to the time-reversal symmetry breaking at the atomic scale. The good description of $FeBr_2$, UO_2 and $ErNi_5$ proves the existence of the discrete atomic-like states both in $3d$, $4f$ and $5f$ systems, on the meV energy scale, confirming *a posteriori* the basic assumption of the crystal-field theory.

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