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The crystal-field and exchange interactions in UGa_2

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Abstract

Magnetic and electronic properties of the hexagonal ferromagnet UGa_2 have been analyzed within the CEF approach in combination with exchange interactions. The $5f^3$ configuration of U^{3+} ions reproduces all experimental observations known, particularly the magnetic moment of $2.7\mu_B$ and Curie temperature T_C of 125 K. The derived electronic structure, with the set of CEF parameters $B_2^0 = +8.38$ K, $B_4^0 = +36.25$ mK, $B_6^0 = -0.5$ mK and $B_6^6 = -52$ mK, yields the state Γ_8 as the charge-formed ground state. Below T_C, strong exchange interactions, relevant to an internal field of 108 T, produce a magnetic singlet ground state. These studies yield strong evidence for strongly localized 5f magnetism of the uranium ion in UGa_2.

UGa_2 crystallizes in a hexagonal crystallographic structure and orders ferromagnetically below T_C of 125 K [1–3]. Magnetic studies on a monocrystalline sample have revealed than the moments are colinear along the a-axis in the hexagonal plane and reaches the value of $2.7\mu_B$/f.u. The paramagnetic susceptibility is largely anisotropic. A Curie–Weiss behaviour observed for the a-axis yields an effective moment of $3.6\mu_B$/U-atom [2].

We have noticed that there is strong similarity in magnetic and electronic (M–E) properties of UGa_2 and $NdNi_5$ compounds [4]. The appearance of magnetic order is shown by a λ-type peak in the specific heat. The spontaneous moment amounts to $2.1\mu_B$ for $NdNi_5$ and $2.7\mu_B$ for UGa_2, much smaller than that for the free trivalent ions. Magnetic properties are highly anisotropic with easy magnetic direction along the a-axis. Anisotropy is also present in the paramagnetic region. The M–E properties of $NdNi_5$ are well described within the crystalline electric field (CEF) approach in combination with exchange (i.e. spin-dependent) interactions by considering the f^3 configuration of the Nd^{3+} ions [4,5]. In this paper we explain the M–E properties of UGa_2 by assuming the $5f^3$ configuration of U^{3+} ions. The lowest multiplet given by Hund's rules is $^4I_{9/2}$ with $S = \frac{3}{2}$, $L = 6$, $J = \frac{9}{2}$ and the Landé factor $g = 8/11$.

The CEF Hamiltonian of hexagonal symmetry for the localized uranium ion can be written as

$$H_{CEF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_6^6 O_6^6. \quad (1)$$

In the paramagnetic region the reciprocal susceptibilities along and perpendicular to the hexagonal c-axis can be written, respectively, as

$$1/\chi_{\parallel} = T/C - n + AB_2^0/C + \Theta(1/T), \quad (2)$$

$$1/\chi_{\perp} = T/C - n - AB_2^0/2C + \Theta^*(1/T), \quad (3)$$

with $A = (2J-1)(2J+3)/5k_B$ and n the molecular field (MF) coefficient between the U-moments. Provided that in sufficiently high temperatures $1/\chi_{\parallel}(T)$ and $1/\chi_{\perp}(T)$ are parallel, the anisotropy of the susceptibility allows the determination of B_2^0. For UGa_2, B_2^0 is estimated to be $+8.38$ K, bigger than that for $NdNi_5$ [4], indicating much stronger CEF interactions in uranium compounds. The joint analysis of the M–E properties leads to the following CEF parameters: $B_2^0 = +8.38$ K, $B_4^0 = +36.25$ mK, $B_6^0 = -0.5$ mK and $B_6^6 = -5.2$ mK, and the MF coefficient $n = 40$ T/μ_B. This set of parameters first of all reproduces a value of T_C of 125 K and an ordered magnetic moment M_U of $2.7\mu_B$/f.u. lying along the a-axis. The temperature variation of the specific heat reproducing a λ-type of peak at T_C (Fig. 1) of M_U and of the paramagnetic susceptibility (Fig. 2), and the field dependence of magnetization along three principal axes of the hexagonal cell (Fig. 3) are in good agreement with the observations. The energy level scheme of the U^{3+} ion, calculated with the present set of CEF parameters is shown in Fig. 4. The CEF interactions split the $^4I_{9/2}$ multiplet into 5 doublets, with overall CEF splitting of 713 K, as the U^{3+} ion with $J = \frac{9}{2}$ is the Kramers ion. Although the second order CEF term B_2^0 is the largest, the higher-order CEF terms make the doublet Γ_8 with the dominant function J_z of $\pm\frac{5}{2}$ as the charge-

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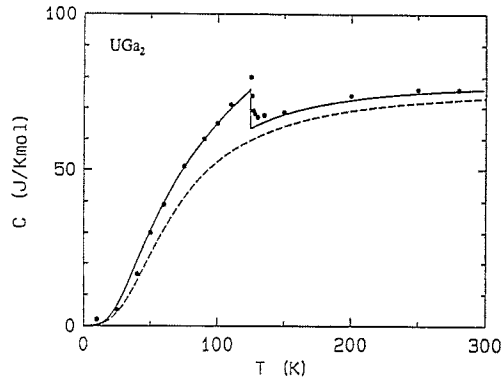

Fig. 1. Calculated specific heat of UGa_2 (solid line). The electronic and phonon contribution (dashed line) is evaluated with a Sommerfeld coefficient $\gamma^e = 5$ mJ/K^2 mol and a Debye temperature $\Theta_D = 280$ K. Points are experimental data taken from Ref. [3].

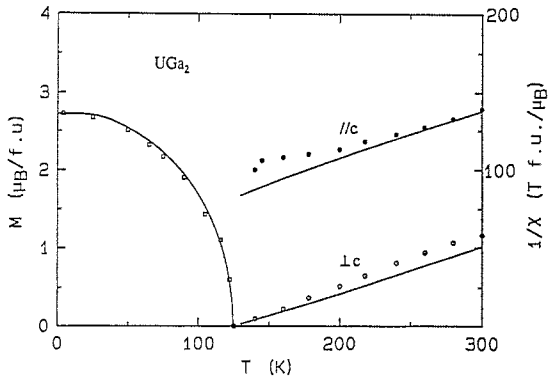

Fig. 2. Calculated temperature variation of the U^{3+} magnetic moment (left scale) and of the reciprocal susceptibility (right scale). Points are data measured for the monocrystalline UGa_2, from Ref. [3].

formed ground state. In the ordered state, all the doublets are split by strong exchange interactions being relevant at 4.2 K to an internal field of 108 T.

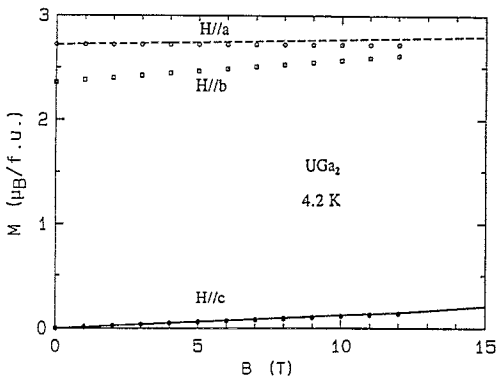

Fig. 3. Calculated field dependence of the U^{3+} magnetic moment for external magnetic fields applied along three principal hexagonal axes in UGa_2. Points are data for the monocrystalline UGa_2 taken from Ref. [3].

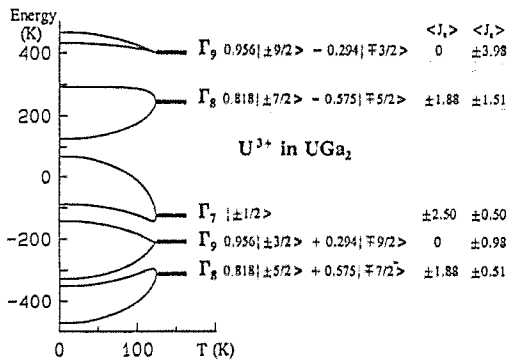

Fig. 4. Energy level scheme of U^{3+} ions ($5f^3$ configuration) in UGa_2 and the appropriate eigenfunctions under the action of the hexagonal CEF interactions. All the CEF levels are Kramers doublets. The exchange interactions split the CEF doublets below 125 K, forming a singlet magnetic state with a moment of $2.7\mu_B$/ion at 4.2 K.

Our analysis is in accordance with photoemission studies [6] which indicate a strong localization of the 5f electrons. It is, however, difficult to distinguish between the $5f^2$ configuration (U^{4+} ions) and the $5f^3$ configuration (U^{3+} ions). These two configurations imply quite comparable values for the effective moment ($3.58\mu_B$ and $3.62\mu_B$, respectively), the saturation moment ($3.27\mu_B$ and $3.20\mu_B$ respectively) and magnetic entropy (19.14 J/K mol and 18.26 J/K mol respectively). Polarized neutron-diffraction studies indicated the preferably 4+ valence [2]. The author could analyze some M–E properties of UGa_2 within the $5f^2$ configuration.

In conclusion, the temperature dependence of the specific heat, magnetization and susceptibility (with distinct anomalies) of UGa_2 have been described by considering the $5f^3$ configuration (U^{3+} ions). It provides strong argument for the localized U-magnetism. The Kramers doublet Γ_8 is the charge-formed ground state, with a largely reduced magnetic moment, giving evidence for the importance of higher-order CEF interactions. The large anisotropy in the M–E properties originates from CEF effects. An expansion of the analysis, including the orthorhombic distortion of the crystal in ferromagnetic phase, is in progress.

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