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Spin and orbital moments in UGa_3

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Abstract

The antiferromagnetic (AF) state of UGa_3 is studied using ^{69}Ga nuclear magnetic resonance. We have observed a drastic change of the hyperfine coupling constant at the Ga site around the AF transition ($T_N = 67$ K) in UGa_3. The internal field at the Ga site appears at slightly lower temperature ∼63 K, indicating another anomaly around 63 K. A possible origin of the anomaly is discussed, in terms of orbital ordering.

1. Introduction

Localized magnetic moments of 5f-electron systems can be understood generally in terms of the Russell–Saunders coupling and Hund's rules. In such a case, the spin and orbital moments can be predicted quantitatively if crystal field levels are known. For itinerant magnets such a picture is no longer applicable, since the spin and orbital moments which appear are usually different from the localized ones. In contrast to the case for 3d itinerant magnets, large orbital moments have been found in 4f and 5f itinerant magnets experimentally [1], although the reasons for this are not well understood. In this context, a contribution of the orbital moment to the magnetism in 4f and 5f itinerant systems is essential.

UGa_3 is an itinerant 5f-electron system with cubic ($AuCu_3$) structure, which shows antiferromagnetic (AF) order ($T_N = 67$ K) with a small ordered moment (∼0.7 μ_B) and a type-II AF structure [2] with propagation vector $\vec{Q} = [\frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$. Another phase transition at 40 K is detected in magnetization measurements. Since the electronic specific heat coefficient $\gamma = 50$ mJ K^{-2} mol^{-1} [3] is relatively large, this compound is regarded as a simple example of a heavy-fermion system with AF order, and has been studied intensively. A previous NMR study [4] has revealed that the direction of the ordered moment is $[11\delta]$ $(1 > \delta > 0)$ for $T_N > T > 40$ K, and that a spin reorientation to $[\alpha, \beta, \gamma]$ $(\alpha, \beta, \gamma \neq)$ occurs at 40 K.

X-ray magnetic scattering results obtained by Mannix *et al* [5] indicate that magnetic scattering appears at slightly lower temperature than T_N (∼62 K), suggesting another transition. In agreement with the x-ray results, our previous NMR study has revealed that hyperfine fields

appear around 63 K, and that this hyperfine field is a consequence of orbital ordering of 5f electrons. On the other hand, recent studies of the magnetic form factor by means of neutron scattering experiments on UGa_3 reveal the existence of a large orbital moment and a drastic change of the spin moment in the AF ordered state [6].

On the basis of these results, we discuss here a possible origin of the anomaly at ∼63 K in terms of orbital and spin moments.

2. Experimental details

The ^{69}Ga NMR has been measured in a single-crystal UGa_3 sample using conventional spin-echo methods. The temperature dependence of the Knight shift ($H \parallel [100]$) has been measured in the paramagnetic and AF states. Since the principal axis of the electric field gradient (EFG) is along [100], two different Ga sites appear when $H \parallel [100]$, i.e., site A with H parallel to the principal axis and site B with H perpendicular to the principal axis.

3. Results and discussion

In the AF state of UGa_3 ($T = 1.5$ K), recent measurements of the magnetic form factor have revealed [6] the existence of large orbital moment μ_L which is antiparallel to the spin moment μ_S as expected from the Hund's third rule ($J = L - S$) for a U^{3+} ion. Due to the effect of itineracy, the ratio μ_L/μ_S is found to be ~ -1.66, which is different from the value -2.56 expected for a U^{3+} free ion. Since the orbital moment is usually small in 3d itinerant systems, the large orbital moment in UGa_3 is considered to be a characteristic of 5f-electron systems. In the paramagnetic state ($T = 80$ K), the local 5f moment seems to disappear and the observed form factor is quite different from that of the AF ordered state but is similar to those of U, UGe_3, and URh_3 [6]. This type of form factor can be interpreted to show that the orbital and spin moments are parallel in the paramagnetic state of a U compound ($J = L + S$), i.e., Hund's third rule is not obeyed [7]. In this picture, the strong change of form factor in the AF state of UGa_3 may correspond to a recovery of Hund's rule. As the form factor has been determined only at 1.5 and 80 K, a behaviour of the recovery is unknown.

If the recovery occurs suddenly, a flop of μ_L against μ_S should occur. This flop may induce a first-order transition in localized insulators since a sudden change of total moment occurs. However, such a recovery in itinerant systems may not be clear, since the magnetic moment can change continuously (crossover) without phase transition. Roughly speaking, the parallel state of L and S is induced in the paramagnetic state when the spin–orbit energy $0.5\xi\chi_s\chi_l$ is less than the orbital Zeeman term $\chi_l\mu_B$ [7]:

$$0.5\xi\chi_s\chi_l < \chi_l\mu_B \tag{1}$$

where χ_s is the spin susceptibility and χ_l is the orbital susceptibility.

Since the spin–orbit coupling energy $\xi = 0.21$ eV for U, the condition $\chi_s < \chi_{sb} \equiv 2\mu_B/\xi = 3.2 \times 10^{-4}$ (emu mol^{-1}) should be satisfied for the parallel state. From a Curie–Weiss fit to the static susceptibility χ in UGa_3 ($H \parallel [100]$) over the temperature range 70–300 K, $\chi \sim 1.5 \times 10^{-3} + 0.0116/(210 + T)$ is obtained. If the observed χ is expressed as $\chi = \chi_s(T) + \chi_l + \chi_{dia}$ ($\chi_{dia} \sim -1 \times 10^{-4}$ (emu mol^{-1}) for UGa_3), the temperature-independent orbital susceptibility χ_l is 1.6×10^{-3}, and thus the spin susceptibility $\chi_s(T)$ is evaluated as $\sim 4 \times 10^{-4}$ (emu mol^{-1}) around 70 K. Since this value is near to the boundary value χ_{sb}, antiparallel-to-parallel recovery is naturally expected in UGa_3. Although it is concluded that the spin susceptibility is negligible in UGa_3 [6], the present results suggest a considerable contribution of spin susceptibility in the paramagnetic state. The observed

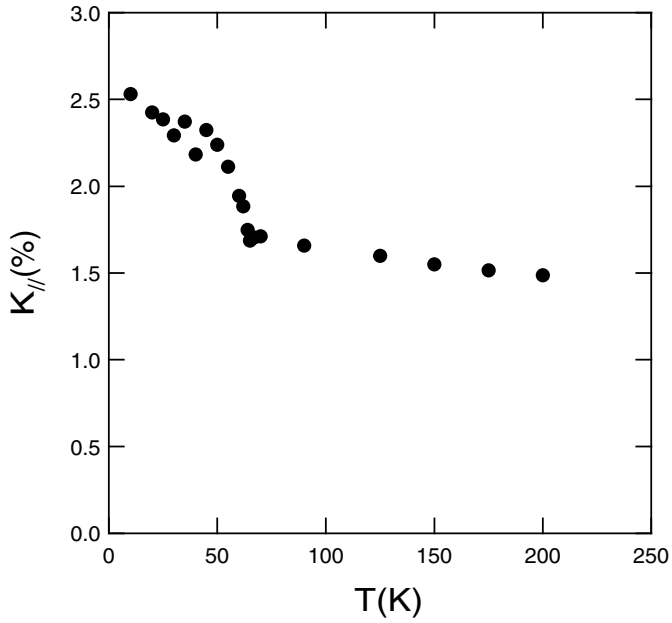

Figure 1. The temperature dependence of the NMR Knight shift at the Ga site (H parallel to the principal axis of the EFG).

temperature-dependent Knight shift also supports this picture, since the T-dependence of the Knight shift is usually due to the T-dependence of the spin susceptibility.

It should be noted that the recovery need not occur at the AF transition. When the system enters the AF ordered state, the effective χ_s may increase, leading to an antiparallel state. In fact, the form factor results suggest a reduction of μ_L/μ_S in the AF ordered state; thus, a pronounced spin–orbit coupling energy is expected. As shown in equation (1), an enhancement of spin–orbit coupling can induce an antiparallel state. One possible scenario for UGa_3 is that the observed orbital ordering around 63 K may be connected with the recovery. In this scenario, orbital ordering appears when the antiparallel state appears with strong spin–orbit interaction energy. Since the spin moment already starts to be ordered at T_N, the enhancement of the spin–orbit coupling favours orbital ordering. Although it is not clear whether the recovery occurs at T_N or ∼63 K, it seems natural to relate the orbital ordering to the strong change of μ_L and μ_S confirmed by the form factor results. Note that no sudden flop with large changes of magnetic moment occurs at $T \sim 63$ K or $T = 40$ K, since no large jump in the specific heat is observed [3].

Figure 1 shows the T-dependence of the Knight shift at site A ($K_\parallel$). In the AF state, an internal field at the Ga site arises from the ordered moment along with the usual Knight shift contribution, which is proportional to the applied field. These two contributions have been evaluated separately. Since the peaks for the B site become broad and weak, the Knight shift at site B ($K_\perp$) could not be determined in the AF ordered state. As shown in the figure, the Knight shift seems to increase drastically around 63 K. Since the T_N of the sample (67 K) is independent of the applied magnetic field up to 9 T, the transition appears at the Ga site at a temperature a few degrees lower, in agreement with the magnetic x-ray scattering results.

The hyperfine coupling constant is obtained using the Knight shift (K) versus susceptibility (χ) plot (figure 2). A good linearity of K–χ is obtained in the paramagnetic and AF states, indicating that the Knight shift evaluated does in fact reflect the static susceptibility at $Q = 0$.

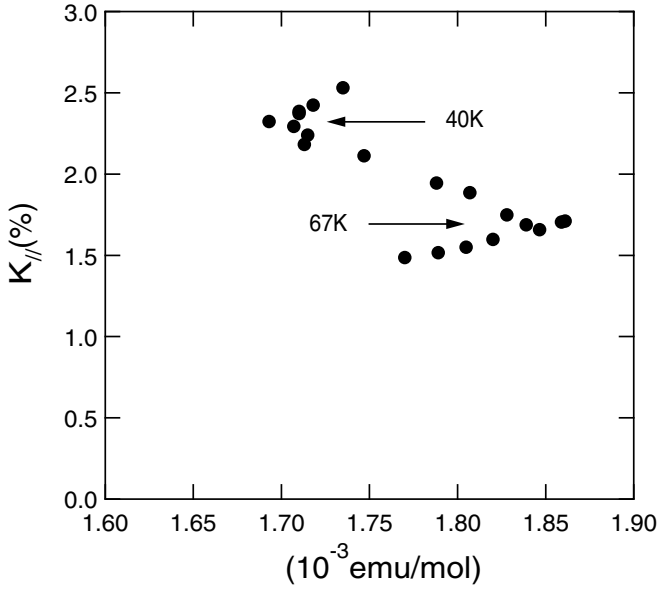

Figure 2. A Knight shift versus static susceptibility (K–χ) plot ($H \parallel [100]$) for the paramagnetic and AF states of UGa_3.

In the paramagnetic state, the hyperfine coupling constant is positive and relatively large. At Ga site a transferred spin polarization of Ga 4p and 4s orbitals from the U moment causes the Knight shift. Since the hyperfine coupling constant is rather isotropic, i.e., $A_\parallel = 140\,\mathrm{kOe}/\mu_B$ and $A_\perp = 120\,\mathrm{kOe}/\mu_B$, the contribution from 4s polarization due to the 5f moment is dominant in the Ga Knight shift.

In the AF state, the value of the hyperfine coupling constant is negative and large, i.e., $A_\parallel = -210\,\mathrm{kOe}/\mu_B$ in UGa_3. Within the present precision of measurements, it is difficult to confirm an anomaly around 63 K. Usually the hyperfine coupling constant at the ligand site is found positive and negative for the cases $J = L - S$ and $J = L + S$, respectively. Since the form factor results suggest the $J = L - S$ case for the AF ordered state, the observed sign of the hyperfine coupling constant is contrary to the usual empirical law. This empirical law is well established, especially for 4f-electron systems. However, in 5f-electron systems, the microscopic origin of the ligand orbital spin polarization is not as simple, since 5f-electron orbitals extend more widely toward the ligand atoms. This leads to direct hybridization between the 5f and ligand orbitals. The sign of the hyperfine coupling constant depends on the ground state of the 5f electrons if the s–5f hybridization is considered [8]. In fact, negative hyperfine coupling constants at ligand sites have been reported for UPt_3 [9] and $U_3Cu_3Sn_4$ [10]. At the very least, the inversion of the hyperfine coupling constant at T_N indicates a drastic change of f-electron state.

The Knight shift versus static susceptibility (K (%) $-$ χ (10^{-3} emu mol^{-1})) relation can be written as

$$K = K_s(T) + K_l = A\chi_s(T) + K_l = 2.5\chi(T) - 2.9 \tag{2}$$

for the paramagnetic state, and

$$K = -3.7\chi(T) + 8.6 \tag{3}$$

for the AF ordered state ($T > 40$ K).

In addition to the drastic change of the hyperfine coupling constant A, the T-independent orbital part K_l is also modified strongly. This is consistent with the strong modification of the orbital moment in the AF state. In other antiferromagnets, e.g., $UPtGa_5$ and $UNiGa_5$ [11], no drastic change of A or K_l has been observed at T_N. Since no orbital ordering or double transition has been observed in these compounds, the drastic change of A and K_l in UGa_3 might be connected with the anomaly at $\sim$63 K. Finally, it should be mentioned that the above discussion on the hyperfine coupling constant is based on an assumption that the hyperfine coupling constant A is independent of T. This assumption is quite reasonable for the paramagnetic state, but if A depends on T in the AF state, a quite different picture can be proposed. We will discuss such a possibility elsewhere.

In conclusion, the temperature dependence of the Knight shift indicates a non-negligible spin susceptibility in UGa_3. The drastic changes of the hyperfine coupling constant and the orbital part of the Knight shift at T_N suggest a strong modification of the orbital and spin moments in the AF state, in agreement with the form factor results. At present, it is not clear whether a real phase transition occurs at $\sim$63 K. Some probe of the dynamics should be carried out around $\sim$63 K in order to clarify the anomaly.

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