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2003 J. Phys.: Condens. Matter 15 S2001

(http://iopscience.iop.org/0953-8984/15/28/314)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) S2001–S2005 PII: S0953-8984(03)62804-3

Ga nuclear magnetic resonance study of $UTGa_5$ (T = Ni, Pt)

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Received 12 November 2002
Published 4 July 2003
Online at stacks.iop.org/JPhysCM/15/S2001

Abstract

Ga nuclear magnetic resonance measurements have been carried out for the 5f antiferromagnets $UNiGa_5$ and $UPtGa_5$. The transferred field at the Ga nuclei has been evaluated. The magnetic structure in the antiferromagnetic region has been confirmed from the microscopic point of view. The mechanism of the hyperfine interaction is discussed.

Recently there has been a great deal of interest in the '115' series of rare-earth and actinide compounds. Following the discovery of unconventional superconductivity in the $CeT'In_5$ series with $T' = Co, Rh, Ir$ [1–3], attention has been drawn to the magnetic properties of the uranium isomorphs $UTGa_5$ (T = Fe, Co, Ni, Ru, Rh, Pd, Os, Ir, Pt), which crystallize in the $HoCoGa_5$-type structure [4]. The U and T atoms form two-dimensional tetragonal lattices in the UGa_3 and TGa_2 layers, respectively. There are two inequivalent Ga sites, namely, Ga(1) at $(1/2, 1/2, 0)$ and Ga(2) at $(1/2, 0, z)$. From the crystallographic point of view, the Ga(2) sites exhibit the C_{2z} symmetry.

In this family, $UNiGa_5$ and $UPtGa_5$ are 5f itinerant antiferromagnets with Néel temperatures $T_N = 86$ K (Ni) and 26 K (Pt), and electronic specific heat coefficients $\gamma = 30\,\mathrm{mJ\,mol^{-1}\,K^{-2}}$ (Ni) and $57\,\mathrm{mJ\,mol^{-1}\,K^{-2}}$ (Pt), respectively [5–7]. Remarkably, recent neutron scattering experiments [8] have revealed that $UNiGa_5$ and $UPtGa_5$ are quite different in magnetic structure below T_N. The magnetic structure of $UNiGa_5$ shows a propagation vector $Q = (1/2, 1/2, 1/2)$, wherein the magnetic moments are ordered antiferromagnetically along the [100] and [001] directions. Meanwhile, Q for $UPtGa_5$ has been determined to be $(0, 0, 1/2)$. This means that the magnetic moments of $UPtGa_5$ are ordered ferromagnetically in the c-plane.

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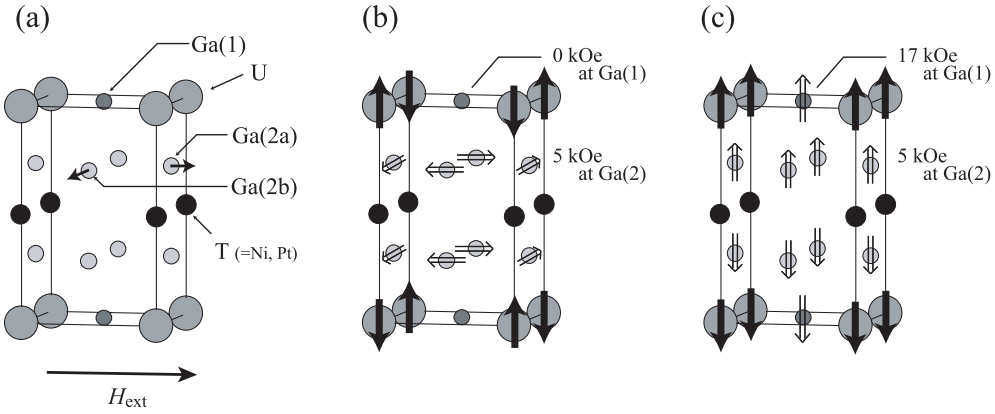

Figure 1. Crystal structure of $UTGa_5$ (a) and schematic view of H_{int} in $UNiGa_5$ (b) and $UPtGa_5$ (c). The thick arrow at the uranium site indicates the direction of the magnetic moments.

In both compounds, it has been suggested that the magnetic moments are aligned along the c-axis.

To examine the magnetic structure of these $UTGa_5$ systems at the microscopic level, we have carried out nuclear magnetic resonance (NMR) measurements on both of them. Just as the magnetic structures of $UNiGa_5$ and $UPtGa_5$ are quite different, so are the NMR spectra below T_N. By evaluating the internal hyperfine fields (HF) at the Ga sites, we have clarified the differences in magnetic structure in a fashion which is complementary to the neutron results [8]. Furthermore, we have found that, in $UNiGa_5$, the direction of the internal field at the Ga(2) sites is perpendicular to the direction of the uranium magnetic moments. The mechanism of the hyperfine interaction is discussed.

Single-crystal samples of $UNiGa_5$ and $UPtGa_5$ prepared by a self-flux method [6, 7] were used. All the measurements were performed using a conventional pulsed NMR spectrometer. In the paramagnetic region, $UNiGa_5$ and $UPtGa_5$ show similar spectra. Since the nuclear spins of ^{69,71}Ga are $\boldsymbol{I} = 3/2$, they undergo quadrupole interactions with their local electrical field gradient (EFG) tensors, as well as Zeeman interactions with the external field H_{ext}. It is to be noted that two 'magnetically' inequivalent Ga(2) sites are observed with $H_{ext} \parallel [100]$, corresponding to the different angles between H_{ext} and the principal axes of the local EFG tensors. As shown in figure 1(a), we define Ga(2a) as the Ga(2) site with the EFG z-axis parallel to H_{ext} (the [100] direction), while Ga(2b) is Ga(2) with the EFG z-axis perpendicular to H_{ext}. From the NMR paramagnetic spectra, we have determined the hyperfine parameters, such as the Knight shift tensors K_α and the quadrupole parameters ν_Q and η for each site in the two compounds [9]. A detailed account of these parameters in the paramagnetic region will be presented elsewhere.

In the antiferromagnetic region, the Ga sites develop the transferred HF fields H_{int} from the magnetic moments. Since H_{int} strongly reflects the orientation of the neighbouring magnetic moments, we can verify the magnetic structure by evaluating H_{int} from the spectrum, as discussed below. The results for H_{int} are summarized in figures 1(b) and (c).

First, we address the NMR spectrum of $UNiGa_5$ with $H_{ext} \parallel [100]$. In the antiferromagnetic-state spectrum shown in figure 2, we find that (1) the Ga(1) resonance lines do not split even in the antiferromagnetic region, although (2) each of the Ga(2a) and Ga(2b) splits into two lines, respectively. Here, (3) the line splittings ΔH of the Ga(2a) and the Ga(2b) are evaluated to be 0.6 and 10.0 kOe, respectively.

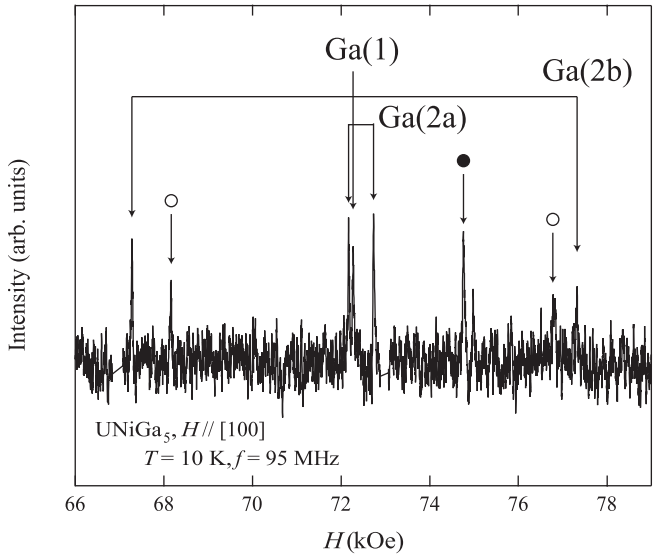

Figure 2. Centre line NMR spectrum of $UNiGa_5$ with $H_{ext} \parallel [100]$ in the antiferromagnetic region. The open and closed circles indicate the satellite lines of Ga(1) and Ga(2b), respectively.

The line splittings of the Ga(2a) and Ga(2b) reflect the antiferromagnetic alignment of H_{int} at the Ga(2) sites, where the parallel/antiparallel component of H_{int} along H_{ext} shifts the resonance position. In the case where $H_{ext} \gg H_{int}$, the magnitude of ΔH can be written $\Delta H = 2H_{int} \cos\theta$, where θ is the angle between H_{ext} and H_{int}.

In considering the line splittings for the Ga(2a) and Ga(2b), the important point is that only one pair of split lines is observed for each the Ga(2) sites, indicating that only one pair of H_{int} fields (having a component parallel/antiparallel to H_{ext}) exists for each site. This implies single-domain structure, maintaining the tetragonal symmetry. If the tetragonal symmetry were to break down in the antiferromagnetic region, the inequivalency of the a and b axes would give rise to two (or more) pairs of H_{int}, which is contrary to the experimental results. Moreover, it is noteworthy that the magnitude of $|H_{int}|$ for Ga(2a) and Ga(2b) should be the same, because the Ga(2a) and Ga(2b) sites are indistinguishable when $H_{ext} = 0$. This means that the difference in ΔH comes from the difference in θ. Because of the tetragonal symmetry, we assume that θ of Ga(2a) is $\pi/2$ different from the θ of Ga(2b). Hence, using the observed ΔH values, the magnitude of $|H_{int}|$ is evaluated to be 5 kOe, and the θ values for Ga(2a) and Ga(2b) are found to be 93.8° and 3.8°, respectively, under the prevailing experimental conditions. Therefore, we conclude that the direction of H_{int} for the Ga(2) is as shown in figure 1(b): H_{int} for the Ga(2a) and Ga(2b) are parallel/antiparallel to the [010] and [100] axes, respectively. With $H_{ext} \parallel [100]$, θ will be 90° and 0° for Ga(2a) and Ga(2b), respectively. The observed deviation of θ from these values is caused by a slight misalignment of H_{ext}. Here, it is to be noted that the direction of H_{int} is perpendicular to the c-axis, which indicates that the C_{2z} symmetry at Ga(2) is broken in the magnetic structure.

In contrast, the fact that no line splitting is observed for Ga(1) clearly indicates that the Ga(1) are not subject to a nonzero H_{int} in the antiferromagnetic region. In fact, in the spectrum with $H_{ext} \parallel [001]$ (not shown), the Ga(1) lines remain unchanged in the antiferromagnetic region. It is concluded that $H_{int} = 0$ for Ga(1).

The fact that H_{int} cancels perfectly at the Ga(1) site, which is located in the centre of a square plaquette of uranium sites, gives strong evidence for antiferromagnetic alignment in

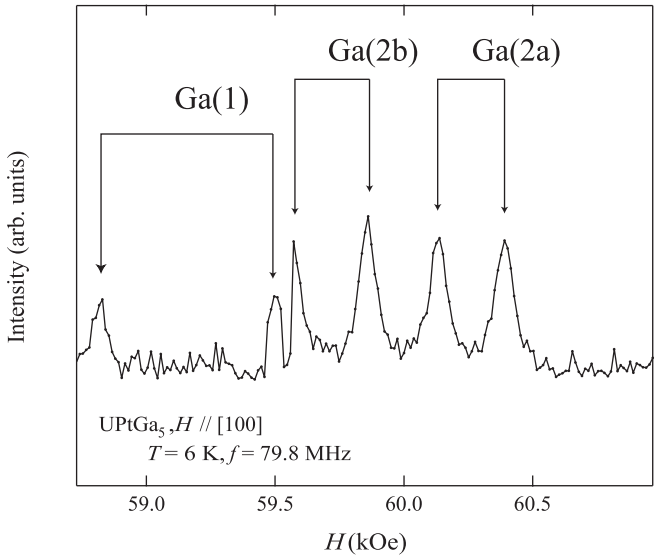

Figure 3. Centre line NMR spectrum of $UPtGa_5$ with $H_{ext} \parallel [100]$ in the antiferromagnetic region. The quadrupolar satellite lines are above and below the field range of this plot.

the square lattice plane. Such magnetic structure also breaks the C_{2z} symmetry at the Ga(2). The symmetric behaviour of H_{int} at the Ga(1) and Ga(2) is consistent with the magnetic structure determined by the neutron diffraction study [8] shown in figure 1(b).

Next, we discuss the NMR spectrum of $UPtGa_5$. The ^{71}Ga centre line spectrum is shown in figure 3. In this case, all resonance lines for the Ga(1), Ga(2a), and Ga(2b) sites are split into pairs of lines in the antiferromagnetic region. ΔH is estimated to be 0.7 kOe for Ga(1) and 0.2 kOe for Ga(2a) and Ga(2b). Our previous zero-applied-field NMR study [10] revealed that the magnitude of $|H_{int}|$ at the Ga(2) site is about 5 kOe, which is considerably larger than the ΔH value for Ga(2) found here. This indicates a vertical orientation of H_{int} at Ga(2a) and Ga(2b) with $H_{ext} \parallel [100]$ perpendicular to it. Because of the misalignment of H_{ext}, H_{int} has a slight parallel/antiparallel component along the direction of H_{ext}, resulting in the line splitting seen in figure 3. The misalignment angle was estimated to be 7° in this experiment. Since H_{int} at the Ga(2a) and Ga(2b) should be identical, H_{int} at Ga(2) is determined to be parallel to the c-axis, in contrast to the case for $UNiGa_5$. The direction of H_{int} preserves the C_{2z} symmetry at Ga(2) magnetically as well as crystallographically.

Concerning the Ga(1), the line splitting seen is due to H_{int}, as in the case of Ga(2) above. Taking into account the fact that only one pair of lines is observed for Ga(1), we can easily rule out an in-plane component of H_{int} at Ga(1). That is, if H_{int} were to have an in-plane component, it would have to lead to a breakdown of the tetragonal symmetry of the magnetic structure. This might be accompanied by a multidomain structure, which is not consistent with our experimental results. Hence, in the case of $UPtGa_5$, it is concluded that $H_{int} \parallel [001]$. Using the ΔH value and the estimated misalignment angle of H_{ext}, we evaluate the magnitude of H_{int} at Ga(1) to be about 17 kOe. These results are summarized in figure 1(c).

The value of H_{int} obtained for $UPtGa_5$ is quite different from that of $UNiGa_5$, reflecting the different magnetic structure. In particular, the direction of H_{int} at Ga(1) in $UPtGa_5$ supports the in-plane ferromagnetic alignment of the magnetic moments. In fact, for the magnetic structure shown in figure 1(c), which was determined from neutron diffraction studies [8], it can easily

be demonstrated that the H_{int} direction at both Ga(1) and Ga(2) should be along the c-axis, based symmetry considerations [11].

Lastly, we note some interesting features of the H_{int} results. In these experiments, we have determined that H_{int} at the Ga(2) in $UNiGa_5$ is perpendicular to the c-axis, i.e. to the direction of the uranium magnetic moments. The orientation for H_{int} can be produced by the dipolar hyperfine interaction. However, for the observed magnetic moment of 0.9 μ_B [8] localized at the uranium site, the simple dipolar value of H_{int} is 1 kOe at most, which is only one-fifth of the observed value.

Of course, the magnetic moment is not necessarily completely localized at the uranium site. Since the 5f electrons are spatially extended, the spin polarization σ is delocalized to some extent. Moreover, the σ of the 5f electrons also give rise to polarization of the conduction electrons, via RKKY interactions. Such delocalization would modify the value of H_{int}. A hybridization effect between Ga-4p and U-5f may also be important in the transverse field. Details will be discussed elsewhere.

In summary, we have compared the spectra of $UNiGa_5$ and $UPtGa_5$ in the antiferromagnetic region and have evaluated the direction and magnitude of H_{int} at the Ga(1) and Ga(2) sites. The magnetic structures have been verified from the microscopic point of view: $UNiGa_5$ shows the antiferromagnetic alignments of the magnetic moments within the planes, while the moments of $UPtGa_5$ are aligned ferromagnetically in the c-plane.

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