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Magnetic field at ^{140}Ce in Dy sites in DyX (X = Cu, Ag) compounds studied by perturbed angular correlation spectroscopy

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Abstract

The magnetic hyperfine field (mhf) at Dy sites in DyAg and DyCu was investigated with perturbed gamma–gamma angular correlation technique using ^{140}La–^{140}Ce probe nuclei. The temperature dependence of the observed mhf shows a deviation from an expected Brillouin-like behavior with a sharp increase of mhf values for temperatures below approximately half of T_N for each compound. This additional magnetic interaction is believed to result from the polarization of Ce spin moments induced by the magnetic field from Dy atoms. The experimental results for the temperature dependence of mhf are fitted by using a model based in the molecular field theory. The results are discussed in terms of the RKKY model for the indirect magnetic coupling between the magnetic Dy ions.

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Keywords: Rare-earth magnetism; Dy-based compounds; Magnetic hyperfine field; Perturbed angular correlation

1. Introduction

The magnetism in DyAg and DyCu is particularly complex where quadrupolar pair interactions yield spontaneous multiaxial (multiple-Q) structures, and multistep metamagnetic processes are observed. DyAg and DyCu order antiferromagnetically, respectively, below 56 K with two additional magnetic transitions at $T_2 = 49$ K and $T_1 = 46.5$ K [1,2], and with $T_N = 62$ K [3]. The crystalline electric field in these compounds induces a threefold axis as easy magnetization direction. The mechanism of spin transfer in such rare-earth compounds, however, is still not well understood. A local investigation of the magnetism in this family of compounds can be very useful to better describe the coupling mechanism between the rare-earth magnetic ions. In this work, perturbed gamma–gamma angular correlation (PAC) technique was used to measure the magnetic hyperfine field B_{hf} in the compounds DyAg and DyCu using the ^{140}La–^{140}Ce nuclear probe at the Dy sites. However, the use of rare-earth probe nuclei in hyperfine interaction techniques introduces a contribution from the probe itself to the magnetic field measured, $B^i_{hf} = B_{4f} + B_{core}$, where B_{4f} is the magnetic field from the 4f moment and B_{core} is the contact field from the s-electrons in the core polarized by the 4f moment. Ce ion has only one 4f electron in the Ce^{3+} state, in the Ce^{4+} state or a mixed valence state.

2. Experimental

The samples were prepared by repeatedly melting the constituent elements, with radioactive ^{140}La substituting about 0.1% of Dy atoms, in an arc furnace under argon atmosphere purified with a hot titanium getterer followed by an annealing at 700 °C in vacuum for 24 h. X-ray diffraction measurement indicated a single phase and the cubic CsCl-type structure with the $Pm\bar{3}m$ space group for the samples. Magnetization measurements were carried out in the temperature range 4–200 K using a superconductor quantum interference device (SQUID).

A conventional fast–slow coincidence set-up with four conical BaF_2 detectors was used for the PAC measurements.

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The well-known 329–487 keV gamma–gamma cascade in ^{140}Ce populated from the beta decay of ^{140}La, with an intermediate level spin $I = 4^+$ at 2083 keV ($T_{1/2} = 3.45$ ns), was used to measure the magnetic hyperfine field (B_{hf}) at Dy sites. The samples were measured in the temperature range of 8–295 K using a closed-cycle helium cryogenic device. The time resolution of the system was about 0.6 ns. A detailed description of the PAC method as well as the experimental procedure can be found elsewhere [4,5]. The experimental data for temperatures below T_N were analyzed for a pure magnetic dipole interaction because the quadrupole moment of the 2083 keV 4^+ state of ^{140}Ce is known to be very small [6], consequently one expects to observe an almost pure magnetic dipole interaction in the magnetic phase of the samples.

3. Results and discussion

The PAC results show well-defined magnetic dipole interaction below the respective magnetic transition temperatures for both compounds. Figs. 1 and 2 show the temperature dependence of the hyperfine field B_{hf} at ^{140}Ce in DyAg and DyCu compounds, respectively. The observed magnetic interaction corresponds to the antiferromagnetic ordering of the Dy moments. The measurements below ~35 K for DyAg and ~40 K for DyCu, however, show a sharp deviation from the expected normal behavior for a simple antiferromagnetic ordering. The ^{140}Ce hyperfine fields, instead of approaching a saturation value, increase sharply at lower temperatures. The measured hyperfine field for DyAg at 33 K is 31.9(9) T while at 8 K it is 79(2) T. The corresponding fields for DyCu are 26.9(6) and 59(1) T, respectively, at 40 and 15 K.

In order to explain the temperature dependence of B_{hf} in both compounds we used a molecular field calculation

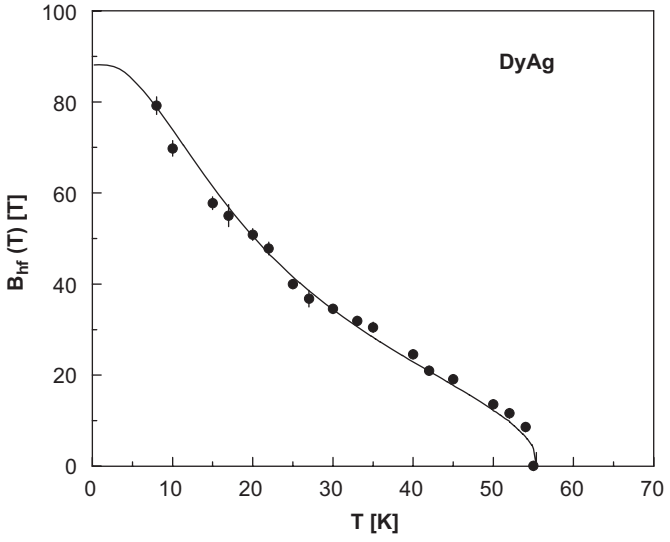

Fig. 1. Temperature dependence of the magnetic hyperfine field at Dy sites in DyAg for ^{140}Ce probe. The solid line represents the fitting to a molecular field model described in the text.

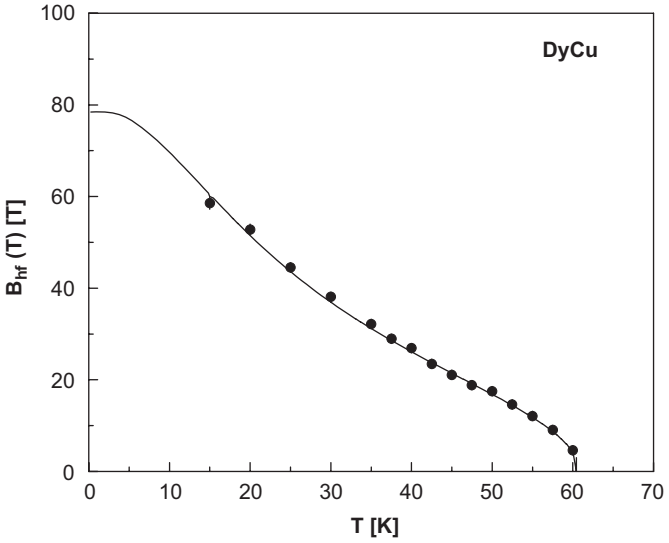

Fig. 2. Temperature dependence of the magnetic hyperfine field at Dy sites in DyCu for ^{140}Ce probe. The solid line represents the fitting to a molecular field model described in the text.

based on a model proposed by Jaccarino et al. [7] and adapted to ^{140}Ce diluted in magnetic rare-earth hosts [8,9]. In this model the effective hyperfine field, B_{hf}, at the probe site is given by the sum of the contributions from the probe ion itself, B^i_{hf} and from the conduction electron polarization, B_{ce}, which scales with the host reduced magnetization $\sigma(T)$. B^i_{hf} is proportional to the thermal average of the impurity moment $\langle J^i \rangle$, which is localized and independent of temperature. The thermal average is taken over its levels in the exchange field of the host B^i_{exc}:

$$B_{hf}(T) = \frac{\langle J^i_z \rangle}{J^i} B^i_{hf}(0) \times B_{J^i}(y) + B_{ce}(0)\sigma(T), \quad (1)$$

where the argument of the Brillouin function $B_{J^i}(y)$ is given by

$$y = -\langle J^i_z \rangle g_{J^i} \mu_B \xi B^i_{exc} + \Delta/kT, \quad (2)$$

where the Landé factor $g_{J^i} = \frac{6}{7}$ and total angular momentum $J^i = \frac{5}{2}$ for Ce^{3+}, and

$$B^i_{exc} = \frac{3kT_C}{(2g_J - 1)(J+1)\mu_B} \times \sigma(T). \quad (3)$$

B^i_{exc} splits the $\frac{5}{2}$ state of $4f^1$ Ce^{3+}, and Δ gives the energy difference of this state relative to the non-magnetic $4f^0$ Ce^{4+} state energy. ξ is a parameter that takes into account the fact that the host–impurity exchange may be different from the host–host exchange. For Dy^{3+} ions, $J = \frac{15}{2}$ and $g_J = \frac{4}{3}$.

The Néel temperature for DyAg and DyCu found in the fitting is 55.7 and 60.9 K, respectively, which agree with the values of 56.6 K [2] and 62 K [10] found in literature.

The issue of interaction between a magnetic impurity ion diluted in a magnetic host is not well understood yet. Particularly, the host–impurity exchange interaction is not well described. The results of the fitting show that this

interaction is stronger for DyCu ($\xi = 0.87$) than DyAg ($\xi = 0.78$). One possible explanation would be the fact that the magnetism in both compounds are affected by other phenomena like strong quadrupolar interactions, multistep metamagnetic process, multiaxial magnetic structures rather than the magnetic interaction directly proportional to the localized magnetic moment at the ion of the host, $\mu = 9.7\,\mu_B$ [1] for DyAg and $\mu = 9.5\,\mu_B$ [11] for DyCu.

The magnetic coupling between rare-earth moments can occur by indirect 4f–4f exchange, which, according to the RKKY theory, is mediated by a spin-polarization of the s-conduction electrons induced by the localized 4f-spins. There is another possibility where the coupling is provided by intra-atomic 4f–5d exchange and direct 5d–5d interaction between the spin polarized 5d-electrons of neighboring rare-earth ions [12]. According to the RKKY theory of indirect coupling the ratio between the conduction electron spin polarization (CEP) and the magnetic ordering temperature is expected to be proportional to $[J_{sf}(g_J - 1)(J + 1)]^{-1}$, where J_{sf} is the s–f coupling constant. Considering the same J_{sf} value for both compounds one expect that the $B_{ce}(0)$ value for DyCu are higher than the value for DyAg. The fitting results to the experimental data show that $B_{ce}(0)$ is 5.4 T for DyAg and 5.8 T for DyCu, which yield a ratio $B_{ce}(0)/T_C$ of 0.0952 for DyAg and 0.0954 for DyCu confirming the RKKY prediction. Furthermore, these values are of the same order of magnitude as the ratio $B_{hf}(0)/T_C = 0.0705$ for DyNiIn compound where the temperature dependence of the magnetic hyperfine field measured with ^{140}Ce at Dy sites [13] does not show any deviation from the Brillouin curve and the main contribution to B_{hf} is supposed to come only from the conduction electron polarization by the Dy ions. Part of the conduction electron polarization comes from the 4f moment of the local Ce ion, besides the polarization from the neighboring magnetic Dy ions. This additional polarization of the conduction electrons present in DyAg and DyCu compounds could explain the higher values $B_{ce}(0)/T_C$ when compared to the value for DyNiIn.

The results of the fitting also show that the values for $B^i_{hf}(0)$ are much higher than $B_{ce}(0)$ and have opposite signs for both compound, -98 and -85 T for DyAg and DyCu, respectively. The explanation for the this contribution to the magnetic interaction is the same as given for the temperature dependence of the B_{hf} at ^{140}Ce in $CeMn_2Ge_2$ [14]. In this compound ^{140}Ce probe nuclei is not an impurity and no local magnetic moment was observed at Ce sites. The local field is believed to result from the polarization of Ce spin moments induced by the magnetic field from Mn moments. In the case of DyAg and DyCu compounds, the magnetic interaction from the impurity is supposed to result from the polarization of Ce f-spin moments induced by the magnetic field from Dy moments. As $B^i_{hf}(0)$ for both compounds is much smaller than the the magnetic hyperfine field of 183 T for the free Ce^{3+} ion [15], it is possible that the ground state of Ce ions has an intermediate valence.

Mössbauer measurements with ^{161}Dy showed that B_{hf} has a constant value of 582 T in the entire range of 4–60 K [16]. This large field is due to the orbital contribution and since the 4f electrons of Dy overlap little with 4f electrons of neighboring Dy, the magnetic interaction is mediated by conduction electrons. As this contribution is much smaller than the orbital contribution, ^{161}Dy probe are not suited to study CEP in DyAg and DyCu compounds. The crystalline field was not taken into account in the calculation of B_{hf}. It may change the relative energy position of the magnetic sublevels J^i_z of Ce^{4+}. First-principles calculations would help to determine the valence of Ce ions as well as the J^i_z ground state.

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