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Magnetic vacancies in antiferromagnetic RAg compounds—A PAC study

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Abstract

Magnetic vacancies have been introduced into the $(\pi,\pi,0)$-type antiferromagnetic structure of cubic RX compounds (R = rare earths, X = Ag, Cu) by replacing magnetic R by non-magnetic Y atoms. The magnetic hyperfine interaction resulting from this break of symmetry at the X-site has been investigated in $R_{1-x}Y_xAg$; R = Gd, Tb, Dy for Y concentrations $0 \leqslant x \leqslant 0.3$ and in $Dy_{0.8}Y_{0.2}Cu$ by perturbed angular correlation (PAC) spectroscopy with ^{111}Cd as probe nucleus. The magnetic hyperfine field produced at ^{111}Cd by one uncompensated nearest neighbor 4f spin is roughly proportional to the spin projection $B_{hf} \sim 0.9\,(g-1)J$ [T]. The temperature dependence of the magnetic hyperfine fields reflects the interaction of the 4f charge distribution with the crystal electric field. For R = Gd and Tb, the fraction of probe nuclei with one nearest Y neighbor is much smaller than expected for a statistical distribution of Y on R sites, suggesting that in these RAg compounds the ^{111}In/^{111}Cd probe atoms favor Ag sites with eight nearest R neighbors.

Keywords: Rare earth intermetallic compounds; Magnetic hyperfine fields; PAC spectroscopy

1. Introduction

Measurements of the magnetic hyperfine interaction at probe nuclei may provide useful information on the properties of magnetically ordered systems. Recently, we have used hyperfine interaction measurements to investigate the coupling mechanism, phase transitions and crystal field effects in rare earth (R) intermetallic compounds RCo_2 [1], R_2In [2] or RAl_2 [3] and an extension to other R intermetallics appear of interest.

An extensively studied group of rare earth intermetallics are the RX compounds with cubic CsCl-type structure, where X is a simple metal [4]. For monovalent X such as Ag or Cu, many of these compounds show antiferromagnetic order below room temperature with a $(\pi,\pi,0)$-type magnetic structure where the 4f-moments in adjacent ferromagnetic (1 1 0) planes are oppositely oriented (e.g. GdAg [5], TbAg, TbCu [6], DyAg [7] and DyCu [8]). In such a structure, all 4f-induced contributions to the magnetic hyperfine field at site X are expected to cancel. A finite magnetic hyperfine interaction arises only on the R site, as recently shown for the probe nucleus ^{140}Ce on the Gd site of GdAg [9].

It is, however, conceivable that information on local magnetic properties at the X-site of RX compounds can be obtained if the antiferromagnetic symmetry is broken by substituting one of the nearest R neighbors of the X-site by a non-magnetic atom such as isovalent yttrium. We have investigated this aspect by studying the magnetic hyperfine interaction in $R_{1-x}Y_xAg$ compounds, using perturbed angular correlation (PAC) spectroscopy [10,11] with ^{111}Cd as probe nucleus. This isotope is populated by the electron capture decay of the 2.8d isotope ^{111}In which according to Ref. [12] is expected to reside on the Ag site.

2. Experimental details

The PAC measurements were carried out with the 171–245 keV cascade of ^{111}Cd. A standard 4-detector

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BaF_2 setup was used, equipped with a closed-cycle He refrigerator for temperature variation in the range $8\,K \leqslant T \leqslant 300\,K$. Samples of $R_{1-x}Y_xAg$ for R = Gd, Tb, Dy, and Er with Y concentrations $0 \leqslant x \leqslant 0.3$ were produced by arc melting of the metallic components in the stoichiometric ratio in an argon atmosphere. Measurements were also performed for DyCu and $Dy_{0.8}Y_{0.2}Cu$. The samples were doped with the PAC probes $^{111}In/^{111}Cd$ by diffusion (800 °C, 12 h) of carrier-free ^{111}In into the host lattices. The CsCl-type structure of the compounds was verified by X-ray diffraction measurements of non-radioactive samples. AC susceptibility measurements, performed in a field $H_{AC} = 1\,Oe$ and frequency of 825 Hz, were consistent with antiferromagnetic order and confirmed the Neel temperatures given in the literature (see Table 1).

3. Results

3.1. RAg compounds

Fig. 1 shows the PAC spectra of ^{111}Cd in paramagnetic GdAg at 300 K and in magnetically ordered GdAg, TbAg, DyAg, and ErAg at temperatures of 8–15 K.

The room temperature (RT) spectra of all RAg and that of DyCu [13] are identical to the one of GdAg shown in Fig. 1. They consist of an almost constant anisotropy, non-modulated in time. This implies the absence of a perturbing quadrupole interaction and confirms that the probe nuclei reside on a site of cubic lattice symmetry, either the R or the Ag site [*Note*: The slight decrease of the anisotropy with time of the RT Spectra can be attributed to lattice imperfections.] The low-temperature spectra of RAg, R = Gd, Dy, and of DyCu [13] are as unperturbed as the spectra at 300 K. Clearly, no magnetic hyperfine field is acting on the probe nuclei in the magnetically ordered phase which identifies the Ag (Cu)-site as the probe site.

Table 1
Parameters of the magnetic hyperfine interaction of ^{111}Cd in $R_{0.8}Y_{0.2}Ag$ compounds and in $Dy_{0.8}Y_{0.2}$ Cu

Compound	f	ν_M^{1Y} (MHz)	$\nu_M^{1Y}/(g-1)J$ (MHz)	T_N (I) T_N (II) (K)	R_{T_N}
$Gd_{0.8}$ $Y_{0.2}Ag$	0.10_2	7.75_6	2.21_2	132 112	0.82
$Tb_{0.8}$ $Y_{0.2}Ag$	0.10_2	6.50_6	2.16_2	101 90	0.85
$Dy_{0.8}$ $Y_{0.2}Ag$	0.30_1	5.20_5	2.08_2	56 45	0.80
$Dy_{0.8}$ $Y_{0.2}Cu$	0.28_2	5.40_5	2.15_2	62 48	0.77

The parameter f describes the fraction of ^{111}Cd probes with one Y nearest neighbour, ν_M^{1Y} is the magnetic frequency of this fraction and $\nu_M^{1Y}/(g-1)J$ the frequency per unit spin. T_N(I) is the Neel temperature of RAg (Cu), taken from Ref. [4]. T_N(II) the Neel temperature of $R_{0.8}Y_{0.2}Ag$(Cu), derived from the ^{111}Cd PAC spectra and R_{T_N} is the ratio of these Neel temperatures $R_{T_N} = T_N(II)/T_N(I)$.

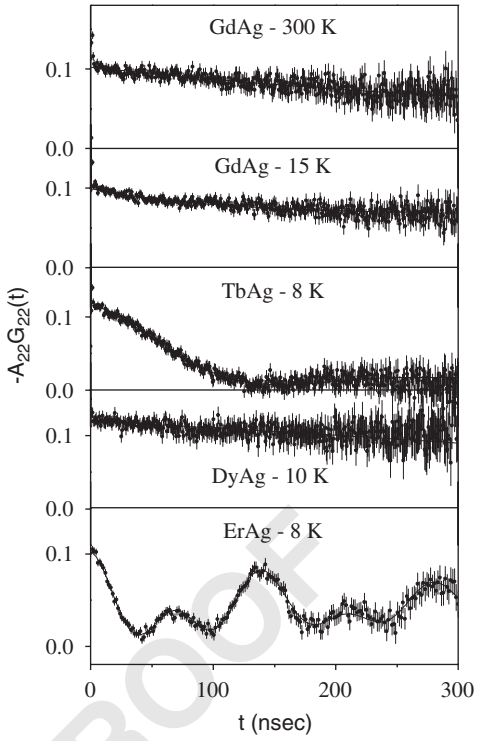

Fig. 1. PAC spectra of ^{111}Cd in GdAg at 300 and 15 K, in TbAg, DyAg and ErAg at 8–10 K.

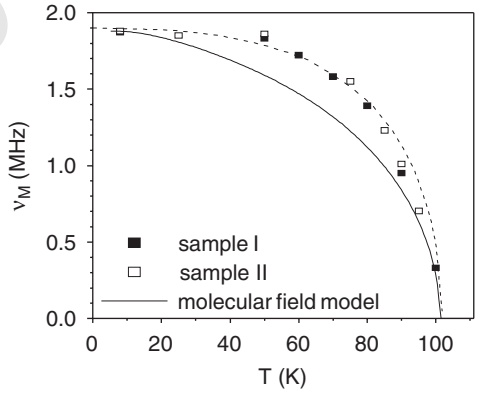

Fig. 2. The temperature dependence of the magnetic interaction frequency of ^{111}Cd in TbAg. The solid line represents the temperature dependence predicted by the molecular field theory; the dotted line connects the experimental data.

In the case of TbAg and ErAg, the low-temperature spectra reflect the presence of a magnetic hyperfine interaction at ^{111}Cd. Figs. 2 and 3 show the temperature dependence of the corresponding magnetic frequencies. The Néel temperatures derived from these data are $T_N = 101$ and 15 K for TbAg and ErAg, respectively, in fair agreement with the values obtained by neutron diffraction [6,14].

It seems rather improbable that the site preference of ^{111}Cd changes with the R constituent. We therefore assume that in TbAg and ErAg ^{111}Cd also occupies the Ag site. The observation of a finite magnetic hyperfine field is then

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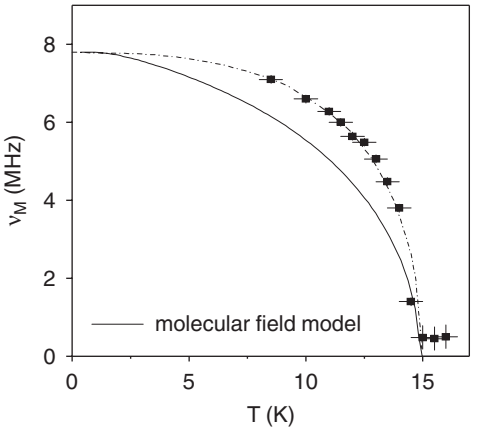

Fig. 3. The temperature dependence of the magnetic interaction frequency of ^{111}Cd in ErAg. The solid line represents the temperature dependence predicted by the molecular field theory; the dotted line connects the experimental data.

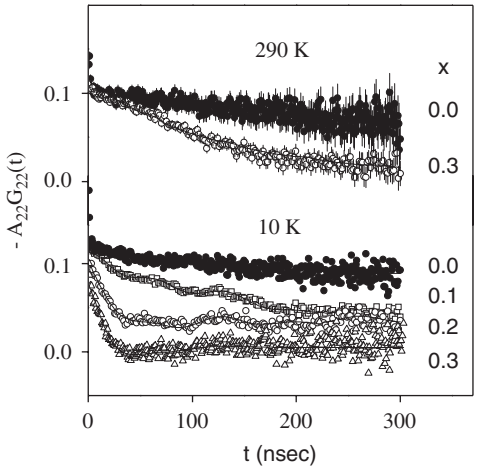

Fig. 4. PAC spectra of ^{111}Cd in $Gd_{1-x}Y_xAg$ at 290 and 10 K for the Y concentrations $x = 0$, 0.1, 0.2 and 0.3.

rather puzzling, at least for TbAg. In the case of ErAg, neutron diffraction [14] suggests the existence of two antiferromagnetic phases, with Neel temperatures of 9.5 and 18 K, respectively. In the range $9.5(5)\,K \leqslant T \leqslant 18(1)\,K$, an incommensurate sinusoidal modulation of the magnetic moments occurs which may give rise to a finite magnetic hyperfine interaction at the Ag site. It cannot be excluded that we slightly underestimate the lowest temperatures, since these were measured with a Si diode at the cold-finger about 2 cm from the sample, and that at nominal 8 K, the sample was actually in the second magnetic phase. In the case of TbAg, the observation of a finite magnetic interaction suggests that the nearest neighbor spin orientation of the probe site differs from the $(\pi,\pi,0)$-type structure, possibly as a result of probe-induced changes of the crystal electric field.

3.2. $R_{1-x}Y_xAg$ compounds

Fig. 4 illustrates the effect, on the ^{111}Cd PAC spectra in the paramagnetic and the antiferromagnetic phase, of the substitution of magnetic R by non-magnetic Y atoms, using $Gd_{1-x}Y_xAg$ as example.

With increasing Y concentration, the decrease of the anisotropy in paramagnetic $Gd_{1-x}Y_xAg$ becomes faster, reflecting increasing quadrupole perturbations related to the increase of chemical disorder. In the antiferromagnetic phase, the spectra for $x > 0$ consist of two components, (i) a monotonous decrease of the anisotropy which is the faster the larger x and (ii) superimposed a small-amplitude oscillation with a period of about 150 ns which is most clearly visible for $x = 0.1$ and 0.2. This oscillation can be attributed to the well-defined magnetic hyperfine field which acts on the probe nuclei when one of the eight nearest R neighbors is replaced by a non-magnetic Y atom. Because of the r^{-3}-dependence of the magnetic hyperfine field, contributions from magnetic vacancies beyond the nearest shell are one order of magnitude smaller. Other configurations with two or more magnetic vacancies in the nearest-neighbor shell give rise to a distribution of magnetic hyperfine fields which leads to the monotonous decrease of the anisotropy. Probes with eight nearest R neighbors are subject to the weak field distribution caused by magnetic vacancies beyond the nearest neighbor shell. The spectra were therefore analyzed by fitting the expression

$$G_{kk}(t) = fG_{kk}^{1Y}(t) + (1-f)G_{kk}^{dist}(t) \quad (1)$$

to the measured data. f is the fraction of probes with one nearest neighbor (n.n.) Y atom, $G_{kk}^{1Y}(t)$, $G_{kk}^{dist}(t)$ are the theoretical perturbation factors for a well-defined magnetic hyperfine field and a field distribution, respectively [10]. In the case $x = 0.1$ and 0.2, the frequency ν_M^{1Y} caused by one n.n. Y atom could be followed up to $T < 100$ K. For $Gd_{0.7}Y_{0.3}Ag$, only the center frequency ν_M^{nY} of the frequency distribution could be determined. The results of the analysis are displayed in Fig. 4, the fraction f and the frequency ν_M^{1Y} at 10 K are listed in Table 1 (Fig. 5).

Similar PAC spectra were observed in $Tb_{0.8}Y_{0.2}Ag$, $Dy_{0.8}Y_{0.2}Ag$ and $Dy_{0.8}Y_{0.2}Cu$ [*Note*: $x = 0.2$ was chosen because at this concentration the largest fraction of probes with 1 n.n. Y is observed]. Fig. 6 compares the spectra of ^{111}Cd in $Dy_{0.8}Y_{0.2}Ag$ to that of ^{111}Cd in DyAg at 10 K. The oscillation caused by one n.n. Y is clearly visible and has larger amplitude than in $Gd_{0.8}Y_{0.2}Ag$. Fraction f and frequency ν_M^{1Y} for $Tb_{0.8}Y_{0.2}Ag$, $Dy_{0.8}Y_{0.2}Ag$ and $Dy_{0.8}Y_{0.2}Cu$ at 10 K are listed in Table 1, the temperature dependences of ν_M^{1Y} and of ν_M^{nY} bfor $Dy_{0.8}Y_{0.2}Ag$ are displayed in Fig. 7.

4. Discussion and summary

^{111}Cd PAC spectroscopy has been used to study the magnetic hyperfine interaction induced by magnetic vacancies at the Ag site of cubic RAg and RCu compounds. In GdAg, DyAg, and DyAg with a $(\pi,\pi,0)$-type antiferromagnetic structure, the hyperfine field con-

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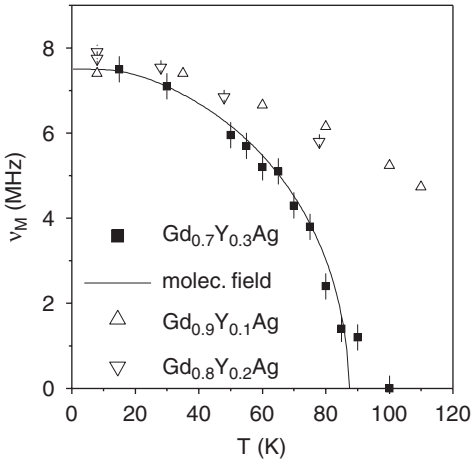

Fig. 5. The temperature dependence of the magnetic interaction frequency of ^{111}Cd in $Gd_{1-x}Y_xAg$ for the Y concentrations $x = 0$, 0.1, 0.2 and 0.3. The open symbols show the frequency ν_M^{1Y}, the full squares correspond to the centre frequency ν_M^{nY} of the frequency distribution. The solid line represents the temperature dependence predicted by the molecular field theory.

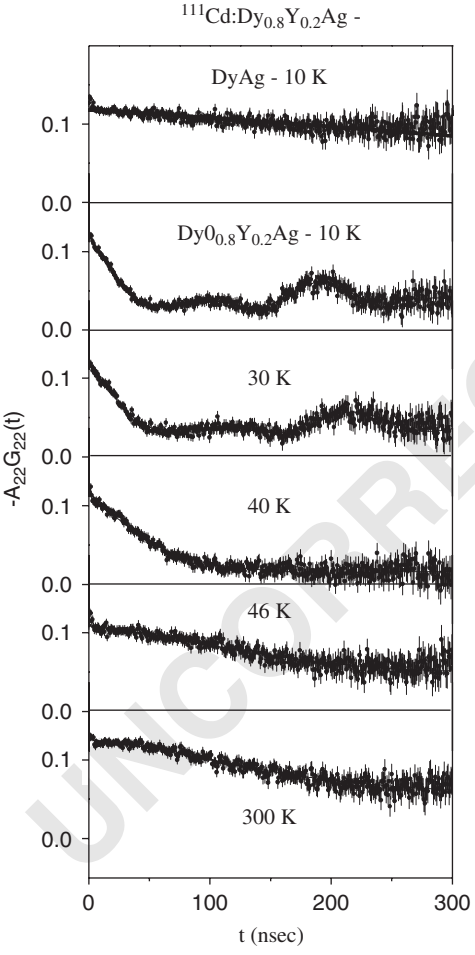

Fig. 6. PAC spectra of ^{111}Cd in DyAg at 10 K and and in $Dy_{0.8}Y_{0.2}Ag$ at temperatures 10 K $\leqslant T \leqslant$ 300 K.

tributions of all 4f spins at the Ag site cancel and the angular correlation in the magnetically ordered phase is unperturbed. In ErAg, the incommensurate sinusoidal

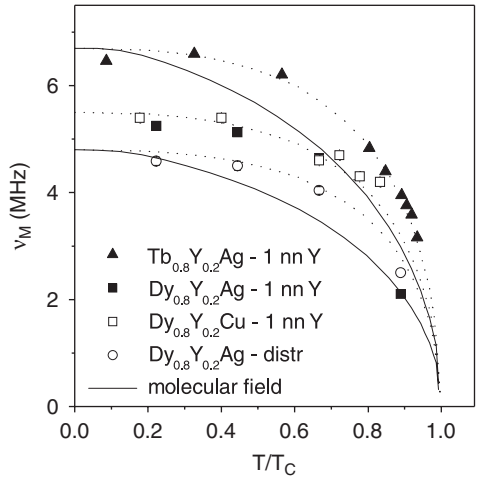

Fig. 7. The temperature dependence of the magnetic interaction frequencies ν_M^{1Y} of ^{111}Cd in $Tb_{0.8}Y_{0.2}Ag$, $Dy_{0.8}Y_{0.2}Ag$, and $Dy_{0.8}Y_{0.2}Cu$, and of ν_M^{nY} for $Dy_{0.8}Y_{0.2}Ag$. The solid line represents the temperature dependence predicted by the molecular field theory; the dotted lines connect the experimental data.

modulation of the magnetic moments leads to a finite magnetic hyperfine field at the Ag site.

A non-magnetic Y atom substituting a nearest rare neighbor of the Ag site breaks the symmetry of the $(\pi,\pi,0)$-type antiferromagnetic structure and produces a magnetic interaction frequency ν_M^{1Y} which—as shown by the almost constant ratio $\nu_M^{1Y}/(g-1)J$ in Table 1 is approximately proportional to the R spin projection $(g-1)J$. The slight decrease of $\nu_M^{1Y}/(g-1)J$ from R = Gd to R = Dy is consistent with a decrease of the effective exchange parameter with increasing R atomic number, as it has been observed in other rare earth intermetallic compounds [2,3]. The frequency ν_M^{1Y} of, e.g. $Gd_{0.8}Y_{0.2}Ag$ corresponds to a magnetic hyperfine field produced by one uncompensated 4f spin of $B_{hf} \sim 3.2$ T. Only a minor part of this value can be explained by the dipolar contribution ($B_{dip} < 0.6$ T for a magnetic moment of $10\mu_B$ at $r_{Ag-R} = 0.31$ nm), the main contribution comes from the polarization of the s-conduction electrons induced by the uncompensated 4f spin.

By breaking the antiferromagnetic symmetry it becomes possible to study the temperature dependence of the magnetic hyperfine interaction and determine the order temperatures. The decrease of the Néel temperature by magnetic dilution is proportional to the Y concentration (see the ratio R_{T_N} of the in Table 1). Figs. 2, 3, 5 and 7 show that only for R = Gd the temperature dependence of the frequency follows the molecular field model. In all other cases, one finds substantial deviations from the corresponding Brillouin function. As Gd is in an s-state, whereas the other R constituents have a non-spherical 4f-charge distribution, this difference suggests a considerable influence of the crystal electric field (CEF) on the temperature dependence of the magnetic hyperfine field. Molecular field calculations including the CEF Hamiltonian for cubic symmetry [15] are under way.

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Assuming a statistical distribution of Y atoms on the R sites, one expects that for $x = 0.2$ about 30 percent of the probes have a magnetic vacancy in the nearest neighbor R shell. Only for the Dy compounds, the experimental value of the fraction f (see Table 1) is consistent with a statistical distribution. For R = Gd and Tb, the fraction of probes with one n.n. Y atom is only 10 percent, suggesting that in these RAg compounds the ^{111}In/^{111}Cd probe atoms favor Ag sites with eight nearest R neighbors.

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