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F. H. M. Cavalcante, L. F. D. Pereira, J. T. Cavalcante, H. Saitovitch, A. W. Carbonari, R. N. Saxena, and M. Forker

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Magnetic hyperfine field in antiferromagnetic RGa_2 (R = Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er) studied by perturbed angular correlation spectroscopy using ^{111}Cd

F. H. M. Cavalcante,[1,2,a)] L. F. D. Pereira,[2] J. T. Cavalcante,[2] H. Saitovitch,[2] A. W. Carbonari,[1] R. N. Saxena,[1] and M. Forker[2,3]

[1]*Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN), São Paulo, Brazil*
[2]*Centro Brasileiro de Pesquisas Físicas (CBPF/MCTI), Rio de Janeiro, Brazil*
[3]*Helmholtz Institut für Strahlen-und Kernphysik-Universität Bonn, Bonn, Germany*

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The magnetic and electric hyperfine interactions of the nuclear probe ^{111}Cd in the hexagonal antiferromagnetic rare earth-gallium RGa_2 (R = Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, and Er) intermetallic compounds have been investigated by perturbed angular correlation (PAC) spectroscopy as a function of temperature. With the exception of R = Nd and Ho, the magnetic hyperfine field B_{hf} is roughly proportional to the spin projection $(g-1)J$ of the R constituent. However, in the group of the light rare earths, the variation of B_{hf} with $(g-1)J$ is much weaker than that for the heavy R constituents, in contrast to the trend reported for all rare earth intermetallics investigated up to now as well as to the trend of the magnetic ordering temperatures of RGa_2. The orientation of the 4*f* spins relative to the c axis of RGa_2 deduced from the angle between B_{hf} and the symmetry axis of the electric field gradient was found to be temperature independent and in agreement with the results of previous magnetization measurements. Except for $SmGa_2$ where the hyperfine field shows an abrupt decrease near T_N, the temperature dependence of $B_{hf}(T)$ is consistent with second order phase transitions. The magnetic ordering temperatures deduced from $B_{hf}(T)$ agree with magnetization and neutron diffraction results. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4795729]

I. INTRODUCTION

Measurements of the magnetic hyperfine field B_{hf} at nuclear sites are a useful source of information on the properties of magnetically ordered compounds. Much of the experimental and theoretical hyperfine interaction work has been focused on magnetic systems involving the rare earth (R) elements Ce to Tm. Among the intermetallic compounds of rare earth with non-magnetic elements studied by hyperfine spectroscopic techniques are phases of the R-Al,[1–3] R-Sn,[4] R-In,[5] R-Ga,[6] and R-Zn (Ref. 7) systems. In the present contribution, we report results of a perturbed angular correlation (PAC) study of the electric and magnetic hyperfine interactions of the nuclear probe ^{111}Cd in the R-Ga intermetallic compounds RGa_2 with R = Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, and Er as a function of temperature (for details on the PAC technique, see Ref. 8). The RGa_2 compounds present antiferromagnetic order below temperatures $T_N < 25$ K. Strong uniaxial magnetic anisotropy and long-range Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction lead to a variety of complex magnetic phenomena[9–11] such as the occurrence of incommensurate magnetic structures and metamagnetic-like transitions in external magnetic fields. RGa_2 crystallize in the hexagonal aluminium-boride (AlB_2) structure (space group *P6/mmm*). The hexagonal symmetry leads an axially symmetric electric field gradient (EFG) at lattice sites. For the ^{111}Cd probe nucleus at Ga sites, this EFG has been determined by Mishra *et al.*[12] through PAC measurements of the nuclear electric quadrupole frequency $\nu_q = eQVzz/h$ in the paramagnetic phase, where Q is the nuclear quadrupole moment and V_{zz} is the principal component of the EFG tensor. In the magnetically ordered phase of hexagonal RGa_2, a magnetic hyperfine field B_{hf} arises and probe nuclei are therefore subject to a combined magnetic and electric hyperfine interaction characterized, in addition to ν_q, by the magnetic hyperfine frequency $\nu_m = g\mu_N B_{hf}/h$ and the angle β between the symmetry axis of the EFG tensor and the orientation of B_{hf}.

II. EXPERIMENTAL PROCEDURE

RGa_2 compounds were synthesized by arc-melting of the metallic constituents with a slight excess (1%–2%) of R elements in Ar atmosphere. For homogeneity, the samples were turned over and remelted several times. Weight losses by arc-melting usually were of the order of 1%–2%. The molten ingots were characterized by X-ray diffraction (XRD) and found to be single phase with the lattice parameters in agreement with the literature.[13] The samples were doped with the PAC probe nucleus $^{111}In \rightarrow ^{111}Cd$ by diffusion in vacuum at 1073 K for 12 h. The PAC measurements were carried out with a standard 4-BaF_2-detector set-up. Temperatures $3.8\,K \le T \le 300\,K$ were reached with a closed-cycle He refrigerator.

a)Electronic mail: fabiohmc@ipen.br

III. RESULTS

Fig. 1 shows the PAC spectra of ^{111}Cd in magnetically ordered RGa_2 at 4 K for the R constituents R = Ce, …, Er and, in the top-most section, typical examples of the spectra observed in the paramagnetic phase at 300 K. The periodic pattern at 300 K is the fingerprint of an axially symmetric quadrupole interaction (QI). The quadrupole frequencies derived from the 300 K spectra agree with those reported by Mishra *et al.*[12] At $T < T_N$, the periodic quadrupole pattern transforms into a non-periodic time dependence of the angular correlation which is characteristic for a combined magnetic and electric hyperfine interaction. The hyperfine interaction (HFI) parameters ν_m, ν_q, and the angle β between the symmetry axis of the EFG tensor and the orientation of B_{hf} derived from the 4 K spectra by a least squares-fit procedure[14] are listed in Table I. In some cases, a satisfactory description of the spectra at $T < T_N$ required the assumption of a second, minority component ($\leq 20\%$) also subject to a combined interaction, which might reflect a strong preference of ^{111}In for other R-Ga phases present in the investigated samples below the limit of XRD detection. The parameters given in Table I refer to the majority component. The orientation of the 4*f* spins relative to the c axis of RGa_2 deduced from β was found to be temperature independent and in agreement with the results of the previous measurements.[10] An example of the temperature variation of the PAC spectra and of the extracted hyperfine field is given in Fig. 2 for $PrGa_2$. The order temperatures derived from the $B_{hf}(T)$ measurements (see Table I) are in fair agreement with the values collected in Ref. 10.

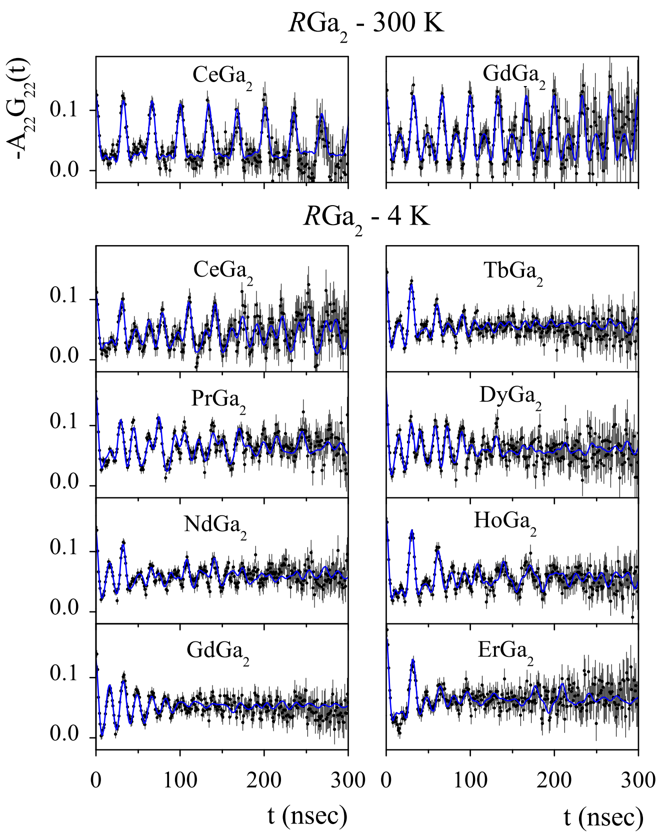

FIG. 1. Perturbed angular correlation spectra of ^{111}Cd in the antiferromagnetic phase of the rare earth-gallium intermetallics RGa_2, R = Ce to Er at 4 K. For comparison, typical spectra of the paramagnetic phase at 300 K are shown in the top-most section.

TABLE I. The hyperfine frequencies $\nu_q = eQV_{ZZ}/h$ and $\nu_m = g\mu_N B_{hf}/h$ of ^{111}Cd in the rare earth-gallium intermetallics RGa_2 at 4 K, derived from the PAC spectra of Fig. 1. In the case of $SmGa_2$, the measurement was carried out at 8 K.[15] β is the angle between the symmetry axis of the EFG tensor and the direction of the magnetic hyperfine field B_{hf}. The precision of β is of the order of 10°. T_N is the magnetic order temperature obtained from the temperature dependence of B_{hf} (precision ~ 1 K). $(g-1)J$ is the projection of the 4*f*-spin on the total angular momentum J, $G = (g-1)^2 J(J+1)$ is the de Gennes factor.

R	$(g-1)J$	ν_m (MHz)	ν_q (MHz)	β (deg)	T_N (K)	$(T_N/G)^{1/2}$ $(K)^{1/2}$
Ce	−0.357	2.6_2	209_2	90	10	7.92
Pr	−0.8	4.3_2	218_2	90	7.0	3.00
Nd	−1.227	12.3_2	208_2	90	9	2.27
Sm	−1.78	7.0_3	222	0	19.5	2.17
Gd	3.5	11.1_5	215_2	90	24	1.22
Tb	3	10.9_2	225_2	90	18.5	1.38
Dy	2.5	8.8_2	231_2	90	11	1.26
Ho	2	1.9_2	215_2	90	7	1.30
Er	1.5	2.0_2	208_2	0	10	1.26

IV. DISCUSSION

In the RKKY theory of magnetic order in rare earth compounds, the indirect coupling between the 4*f*-electrons is mediated by the *s*-conduction electrons, which are spin polarized by exchange with the 4*f*-electrons. This spin polarization leads, via the Fermi contact term in the nucleus-electron interaction, to a magnetic hyperfine field at the nuclei of non-rare-earth atoms. For closed shell nuclear probes such as ^{111}Cd, B_{hf} can therefore be expected to be proportional to the spin polarization which in the free-electron RKKY theory leads to[16]

$$B_{hf} \propto \Gamma\langle S_z\rangle \sum_{n\neq m} F(2k_F R_{nm}) = \Gamma(g-1)J \sum_{n\neq m} F(2k_F R_{nm}). \tag{1}$$

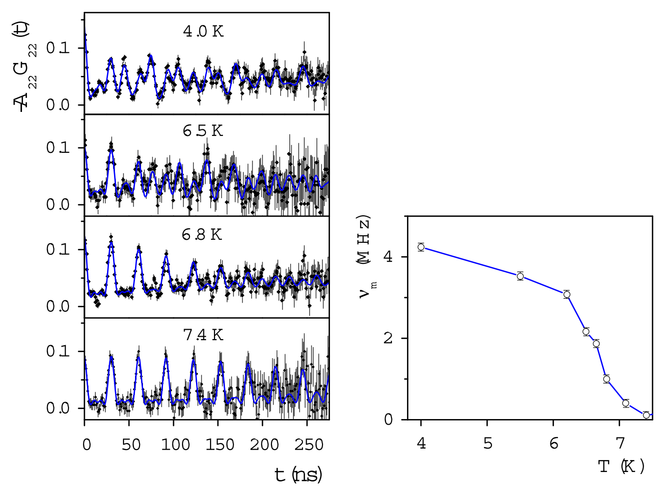

FIG. 2. PAC spectra of ^{111}Cd in $PrGa_2$ and the extracted magnetic hyperfine frequency ν_m (right-hand section) as a function of temperature.

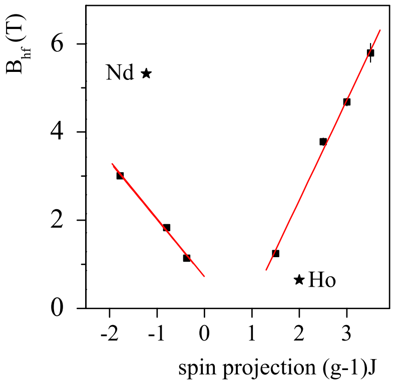

FIG. 3. The spin dependence of the magnetic hyperfine field of ^{111}Cd in RGa_2. $NdGa_2$ and $HoGa_2$ (marked by asterisks) strongly deviate from the systematic variation shown by the other RGa_2.

Here, Γ is an effective *s-f* exchange constant, $(g-1)J$ is the spin projection, and F(x) is the oscillating RKKY function. In the case of RGa_2, the RKKY function varies little with the R constituent[9] and one might therefore expect $B_{hf} \propto \Gamma(g-1)J$. The plot of the experimental hyperfine field values versus the spin projection in Fig. 3 reflects a rough overall proportionality between B_{hf} and $(g-1)J$ with two unexpected features: (i) The values for R = Nd and Ho strongly deviate from the trends of the light R (LR) and the heavy R (HR) constituents. (ii) The slope of B_{hf} versus $(g-1)J$ of the HR group $[B_{hf} = 5.27(24)(g-1)J - 4.8(7)]$ is steeper than that of the LR group $[B_{hf} = 3.04(7)(g-1)J - 1.6(3)]$ (excluding R = Ho, Nd). If this is attributed to different exchange constants of the HR and the LR group, one arrives at a ratio $\Gamma_{LR}/\Gamma_{HR} \sim 0.6$. These observations for RGa_2 strongly differ from those reported for practically all other rare intermetallics investigated up to now.[5,6,17,18] Usually, the variation of B_{hf} with $(g-1)J$ is stronger in the LR group with a ratio $\Gamma_{LR}/\Gamma_{HR} \sim 1.5$. The larger exchange parameter in the LR group suggested by most hyperfine field measurements has been related by Delyagin and Krylov[6] to the higher degree of 4*f*-5*d* overlap suggested by the radial extensions of the 4*f* and 5*d* electrons in the first and the second halves of the R series. It is interesting to note that in contrast to the spin dependence of B_{hf}, the variation of the order temperatures of RGa_2 obeys the de Gennes relation $T_N \propto \Gamma^2(g-1)^2J(J+1) = \Gamma^2 G$ with $\Gamma_{LR} > \Gamma_{HR}$. From the ratios $(T_N/G)^{1/2}$ listed in the last column of Table I (excepting R = Ce), one obtains $\Gamma_{LR}/\Gamma_{HR} \sim 1.9(4)$. This remarkable difference in the trends of B_{hf} and T_N possibly reflects local changes of the spin polarization related to the impurity nature of the PAC probe ^{111}In/^{111}Cd. For a test of this hypothesis, measurements of B_{hf} with other probe nuclei are being prepared.

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