2005 International Nuclear Atlantic Conference - INAC 2005
Santos, SP, Brazil, August 28 to September 2, 2005
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: 85-99141-01-5

INVESTIGATION OF THE LOCAL MAGNETISM AT RARE-EARTH SITES IN RAg (R = RARE-EARTH) INTERMETALLIC COMPOUNDS USING PERTURBED GAMMA-GAMMA ANGULAR CORRELATION SPECTROSCOPY

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ABSTRACT

Time differential perturbed gamma-gamma angular correlation (TDPAC) technique was used to measure the magnetic hyperfine field (mhf) at rare earth sites in intermetallic compounds RAg (R = Dy, Gd, Ho, Nd, Tb) in order to study the behavior of the mhf with 4f-electron density variation. ^{140}Ce was used as probe nuclei in the TDPAC measurements, which was carried out in the temperature range of 10 K to 300 K. The results of the temperature dependence of the mhf show a standard antiferromagnetic behavior below the magnetic order temperature but, for lower temperatures, it was observed a sharp deviation from the expected Brillouin-like behavior with an increase of the mhf values. The values of the mhf for each compound at the lowest temperature just above the deviation were compared to the respective projection of the rare-earth spin on the total angular momentum J. The results showed to follow a similar behavior when compared to those for the magnetic moment of the compounds.

1. INTRODUCTION

In recent decades, many efforts have been made on the investigation of magnetism in compounds with rare-earth (R) elements due to the possibility of technological application in industry (laser, luminescent materials, permanent magnets, glasses) and medicine (contrast agents) [1], as well as the interest in basic scientific research, because of its electric and magnetic properties and its non-usual structures. On the other hand, the study of the origin of magnetism and formation of magnetic moments in rare earth atoms in such compounds, as well as their interactions with neighbors atoms have become one of the main objectives of basic research in magnetism. Therefore, a systematic microscopic investigation in these compounds, where rare earth elements varies from La to Lu, is of great interest, because it can provide information on the origin of magnetic interactions.

Magnetic properties in substances have origin in the magnetic moment of the electrons and its atoms. Electrons present two types of movement: orbital and spin, each one possessing an associated magnetic moment. The magnetic moment of the ion is then originated in the magnetic moments and spins of each electron. Compounds where one of the constituent is a rare-earth ion are expected to present high values of the orbital magnetic moments due to incomplete 4f layer. What distinguishes the magnetic behavior of rare-earth compounds is the number of unpaired electrons in the internal 4f layer with radius of approximately 0,3 Å [2].

The compounds studied in this have chemical formula RX with a CsCl prototype crystal structure, the cubic body-centered structure (BCC) shown in Fig. 1.

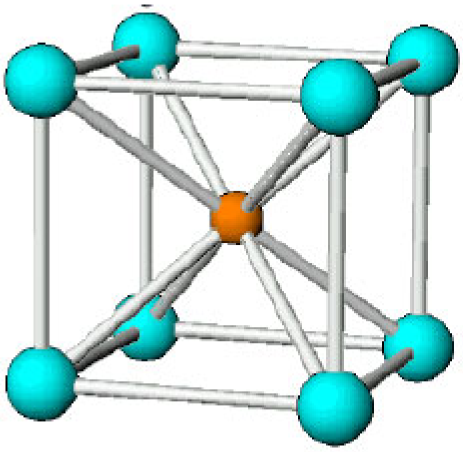

FIGURE 1 - CsCl structure

In general, intermetallic compounds of the RX type, where X is a monovalent ion like Ag, Cu, Zn, Cd present interesting magnetic properties [3-6]. These compounds have an antiferromagnetic ordering below transition temperature [7,8]. The magnetic behavior is determined by a partially full 4f layer, where local moments interact by means of the RKKY indirect exchange mediated by conduction electrons (5d6s), where the electrons 5d has an important paper in the process of magnetic interaction. The antiferromagnetic (π, π, 0) structure in these compounds is built up by ferromagnetic (1,1,0) planes coupled antiferromagnetically. The intermetallic behavior in these compounds may be complex due to the interactions of several parameters, as the possibility of existing incommensurable magnetic vectors, quadrupole interactions, crystalline field splitting and even spin fluctuation near Néel temperature.

In the present work we investigated the mhf at R sites in intermetallic compounds of the type RAg (R = Gd, Tb, Dy, Ho), as a function of the number of 4f electrons. These compounds crystallize in a same crystalline structure, which makes easier the comparison of the mhf from the f band.

In such compounds there is the possibility of hybridization of 4f states in R as well as 4d states in Ag. Although the distance between 4f core electrons and local 4d electrons in Ag seem to be high enough to exclude hybridization, that effect affects spin moments of 4f layer [9].

Hyperfine interaction techniques, especially Time Differential Perturbed Angular Correlation (TDPAC) technique, have proven to be adequate in the study of magnetism in rare-earth compounds, due to its local character of investigation, because it can measure hyperfine magnetic fields (CHM) directly in a specific site inside the material. CHM can “feel” long range interactions of RKKY type, mediated by conduction electrons as well as orbital contributions due to ion valence electrons. The TDPAC can also supply information about the direction of magnetic moments in these compounds. The TDPAC method is based on the observation of hyperfine interaction of nuclear moments with extra-nuclear magnetic field or electric As any technique, TDPAC has its peculiarities and demands. In this case, it is necessary to chemically introduce the probe nuclei into the sample. However, the number of probe nuclei (approximately 10^{11}) required is several orders of magnitude smaller than the number of atoms in the sample. This advantage of TDPAC is very important because the interference in the properties of the sample under study. The probe nuclei act as "observers" sending information about interactions in their next neighborhood, via gamma-ray emission.

In the TDPAC experiment the probes must emit a cascade of gamma rays with an intermediate state with a life time of the order of nanoseconds. The most commonly used probe nuclei are: ^{111}Cd (from the decay of ^{111}In or ^{111}mCd), ^{181}Ta (^{181}Hf), ^{100}Rh (^{100}Pd), ^{140}Ce (^{140}La), ^{99}Ru (^{99}Rh). Further information about the method can be find in reference 10.

2. EXPERIMENTAL

Samples of RAg were prepared by repeatedly melting the constituent elements (R 99.99% and Ag 99.9985%) under argon atmosphere purified with a hot titanium getter in an arc furnace. The samples used for TDPAC measurements were prepared in a similar way but with radioactive ^{140}La (obtained by neutron irradiation of lanthanum metal) substituting about 0.1% of R atoms. Samples were annealed under an atmosphere of ultra pure Ar for 48 h at 700 ºC.

The structure of the samples was checked by X-ray diffraction, which indicated a single phase and the cubic CsCl-type structure with the $Pm\overline{3}m$ space group. Magnetization measurements were carried out in the temperature range of 4-200 K using a superconductor quantum interference device (SQUID).

The TDPAC measurements were carried out with a conventional fast-slow coincidence set-up with four conical BaF_2 detectors. The gamma cascade of 329-487 keV populated from the decay of ^{140}La with an intermediate level with spin $I = 4^+$ at 2083 keV ($T_{1/2} = 3.45$ ns) in ^{140}Ce was used to measure the magnetic hyperfine field (*mhf*) at R sites. The samples were measured in the temperature range of 10-295 K by using a closed-cycle helium cryogenic device. The time resolution of the system was about 0.6 ns for the ^{140}Ce gamma cascade.

The perturbation factor $G_{22}(t)$ of the correlation function contains detailed information about the hyperfine interaction. Measurement of $G_{22}(t)$ allows the determination of the Larmor frequency $\omega_L = \mu_N g B_{hf} / \hbar$, from the known g-factor of the 2083 keV state of ^{140}Ce, and consequent determination of the magnetic hyperfine field B_{hf}. The experimental data for temperatures below T_N were analyzed for a pure magnetic dipole interaction.

3. RESULTS AND DISCUSSION

The X-Ray diffraction spectrum with a Rietveld analysis for the GdAg compound is shown in figure 2. In this picture, the solid curve is the Rietveld fitting to the experimental data and the curve in the bottom is the difference between experimental and fitted curves. The results for the lattice parameters are in agreement with those from literature. The broadening of the peaks in the X-ray spectrum may be caused by a lattice distortion during the arc melting of the samples. Although samples were annealed, apparently it was not enough to eliminate the lattice stress. Figure 3 shows the magnetization versus temperature for the GdAg compound.

The magnetization shows a standard behavior with an increase in the values from 200 K to around 130 K, then a slight decrease followed by a complex behavior for smaller temperatures. The Néel temperature was determined to be 130 K, which agrees with the values from literature. The influence of the 4f spin density on the mhf (B_{HF}) is plotted in figure 4 along with the correspondent values for the magnetic moment of each compound.

From the figure we can observe a similar behavior between the B_{hf} and the magnetic moment values with the projection of the R spin.

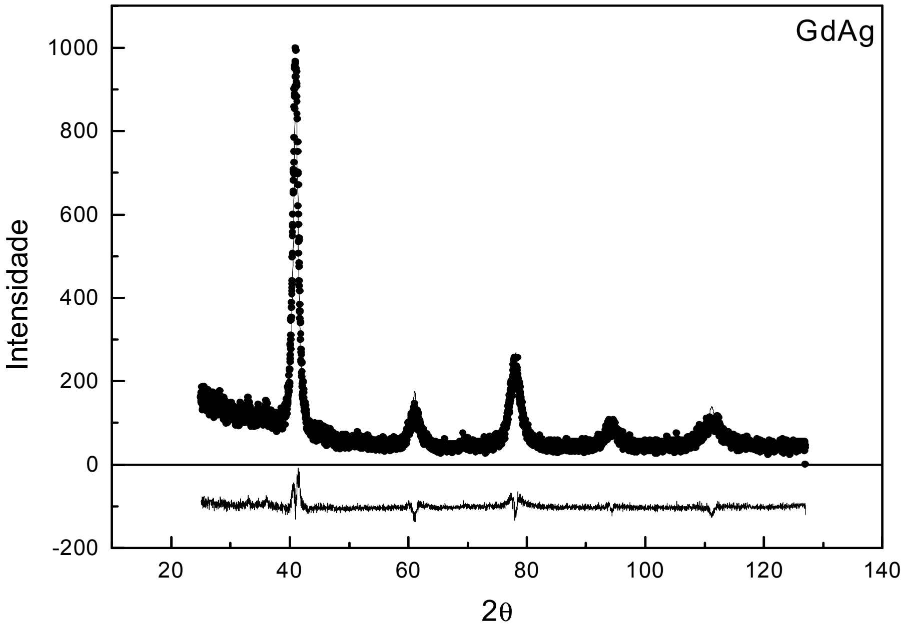

Figure 2. X-Ray spectra for GdAg

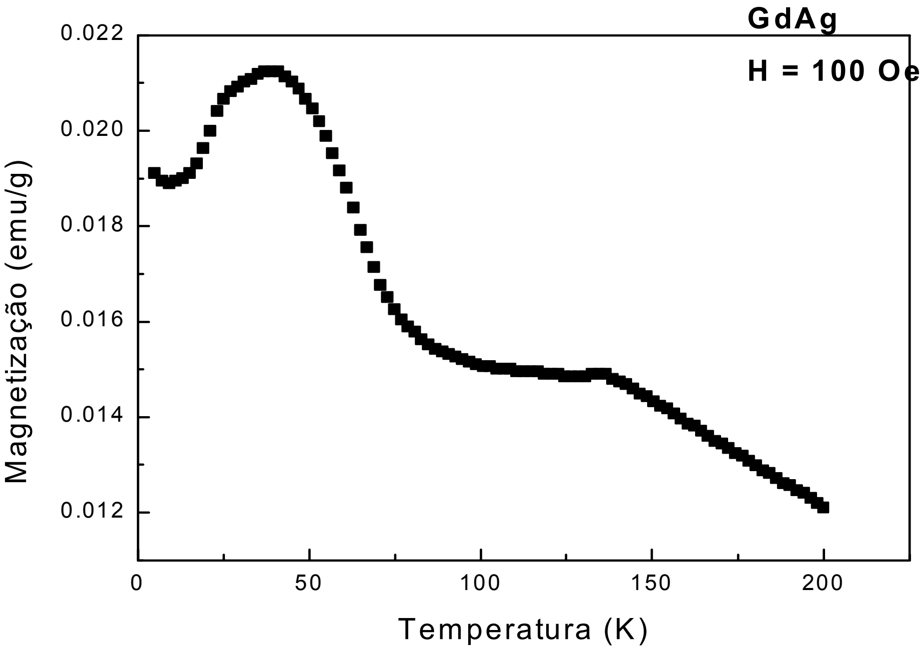

Figure 3. Magnetization Squid spectra for GdAg

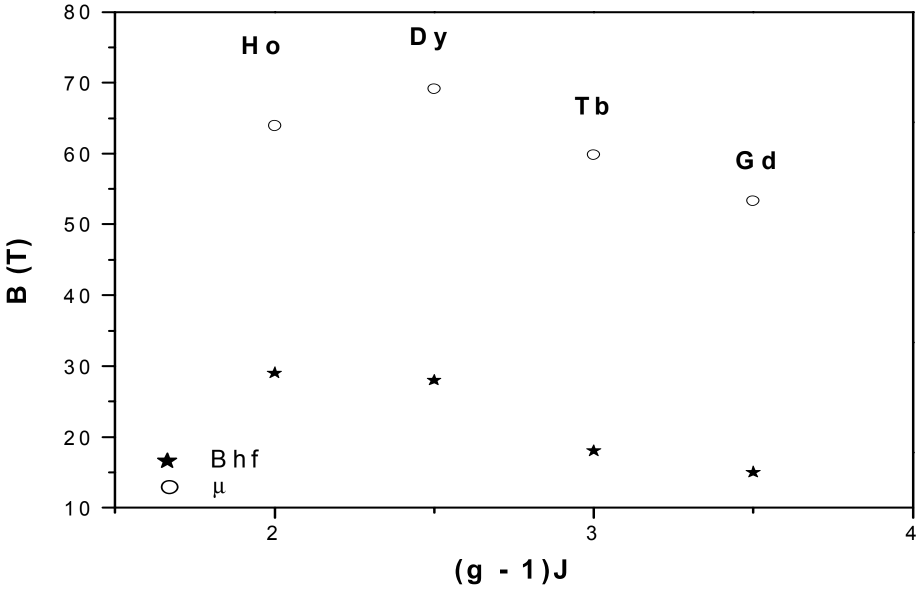

Figure 3.The 4f spin dependence of the magnetic hyperfine field B_{HF} of ^{140}Ce in RAg across the series o f RAg.

ACKNOWLEDGMENTS

Partial support for this research was provided by the Fundação de Amparo para Pesquisa do Estado de São Paulo (FAPESP). Authors FHMC and AWC thankfully acknowledges the support provided by CNPq in the form of a research fellowship.

REFERENCES

1. T. S. Martins and P. C. Isolani, *Química Nova*, v. 28, n. 1, p. 111-117, 2005.
2. C. Kittel, *Introdução a Física do Estado Sólido*. Guanabara Dois.1978.
3. C. Chao; L. Luo; P. Duwez, *J. Applied Phys*., v. 34, n. 7, p. 1971-1973, 1963.
4. R. E. Walline; W. E. Wallace; *The J. Chem. Phys*., v. 41, n. 11, p. 3285-3288, 1964.
5. K. Sekizawa; K. Yasukochi, *J. of the Physical Society of Japan*, v. 21, n. 4, p. 684, 1966.
6. E. Burzo; I. Urzu; Pierre, *J., Phys Stat. Sol. (b)*, v. 51, p. 4637, 1972.
7. U. Kober; W. Kinzel; W. Zinn, *J. Magn. Magn. Mater*., v. 25, p. 124, 1981.
8. K. Yagasaki; Y. Hidaka; H. Fujii; T. Okamoto, *J. Phys. Soc. Jpn*., v. 45, p. 110, 1978.
9. J. Szade; M. Neumann, *J. Phys. Condens. Matter*, v. 11, p. 3887-3896, 1999.
10. W. Pendl Jr., R.N. Saxena, A.W. Carbonari, J. Mestnik-Filho J. Schaft, *J. Phys.: Condens. Matter*. v. 8, p. 11317, 1996.