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Temperature Dependence of the Hyperfine Magnetic Field at ^{140}Ce in Orthorhombic Tb_3In_5

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Abstract. Time differential perturbed γ-γ angular correlation technique was used to measure the magnetic hyperfine field (MHF) at Tb sites in the intermetallic compound Tb_3In_5 using the $^{140}La \rightarrow {}^{140}Ce$ nuclear probe. The measurements were carried out in the temperature range of 8 to 295 K. Two different temperature dependent magnetic frequencies were observed below 30 K, which were assigned as ^{140}Ce substituting the two inequivalent Tb sites in the orthorhombic structure of Tb_3In_5. The temperature dependence of MHF also shows a possible deviation from an expected Brillouin-like behavior for temperatures below 18 K. A Néel transition at 27 K was observed from magnetization measurements in the samples. The magnetization as a function of the applied magnetic field was measured at two temperatures, 5 and 40 K, and the results show anti-ferromagnetic and a typical paramagnetic behavior, respectively. In both cases it was not observed saturation under high magnetic field.

Key Words: ^{140}Ce probe, magnetic hyperfine field, PAC spectroscopy, rare earth magnetism.

1. Introduction

The investigation of the local magnetism in rare earth compounds is very important to understand the rich variety of complex phenomena, which occur in such compounds like Kondo effect, heavy fermion behavior, mixed valence, etc. The origin of the magnetism in these compounds is due to the strongly localized magnetic moment, which is associated to 4f electrons of rare earth atoms. In many compounds, the 4f electrons can interact with other electrons via hybridization with, for instance, s electrons, and lose part of their localized magnetic moment. The investigation of the magnetic hyperfine field (MHF) in rare earth compounds is, therefore important because it is a measurement of the magnetic field in the neighborhood of a particular site and can be associated to magnetic moments in this site or at nearest neighbor sites. The magnetic and electric hyperfine interaction at ^{111}Cd impurity using the Time differential perturbed γ-γ angular correlation (TDPAC) technique has been systematically

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used to investigate intermetallic compounds R_xA_y (R = rare earth element, and A = non-magnetic metal), in the particular those with A = In [1–3], and A = Al [4]. The purpose of such a systematic investigation is to better understand the coupling mechanism that is responsible for the rare earth magnetism. As a part of this study, the intermetallic compound Tb_3In_5 has been investigated ^{111}Cd and the results for previous TDPAC measurements using as probe nuclei were reported [5].

The Tb_3In_5 compound crystallizes in the orthorhombic Pu_3Pd_5-type structure [6, 7], where Tb and In atoms can occupy two and three structurally different sites, respectively. This complex structure makes the local investigation of Tb_3In_5 be quite difficult due to the presence of a complex quadrupole interaction. In the previous measurements [5] with ^{111}Cd, the TDPAC spectrum at 10 K showed a time dependent attenuation of the anisotropy when compared with that obtained at room temperature. This fact was interpreted as due to the presence of the magnetic interaction. However, It was not possible to fit a function that takes into account a combined magnetic and electric interaction. The computer program used to perform this fitting is based on the calculation of Böstrom *et al.* [8], and it is adequate when the electric field gradient (EFG) is axially symmetric ($\eta = 0$). When the asymmetry parameter is small, the program is still adequate if the Larmor frequency (ω_L) of the magnetic interaction is greater than the quadrupole frequency (ω_Q) of the electric interaction. In the case of measurements using ^{111}Cd, the asymmetric electric interaction was very strong and, possibly much larger than the magnetic interaction. Consequently, it was not possible to obtain any information about the magnetic hyperfine field for the Tb_3In_5 compound. The quadrupole moment of the 2083 keV 4^+ state of ^{140}Ce, used as probe nuclei in the present work, is known to be very small [9], and one expects to observe an almost pure magnetic dipole interaction in the TDPAC measurements, which opens the possibility to investigate the local magnetic field in the Tb_3In_5 compound.

2. Experimental

The samples were prepared by repeated arc melting the constituent elements in arc furnace under argon atmosphere purified with a hot titanium getterer. Radioactive ^{140}La nuclei (obtained by neutron irradiation of lanthanum metal in the IEA-R1 research reactor at IPEN) substituting about 0.1% of Tb atoms were melted along with the constituent elements. Samples were annealed at 900°C in a pressure of 1 atm of ultra-pure Ar. Crystal structures of the samples could not be checked by powder X-ray-diffraction measurements due to the plastic deformation during the powdering process [5].

As Tb_3In_5 compound oxidizes very quickly, a second sample, without the radioactive ^{140}La, was prepared for the magnetization measurements. These measurements were carried out in a superconductor quantum interference device

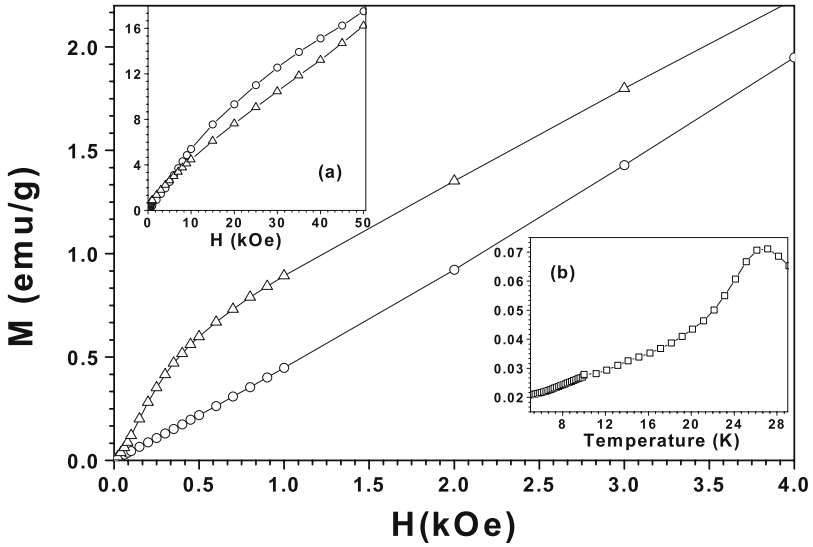

Figure 1. Magnetization curves as a function of applied magnetic field performed at $T = 5$ K (○) and 40 K (△). The insets show (a) in the full interval of H, and (b) the temperature dependence of the magnetization at $H = 1.0$ kOe.

(SQUID) in the temperature range of 5 to 30 K with a rate of 1 K/min and an applied magnetic field of 1 kOe. In addition, the magnetization was also measured as a function of the applied field for 5 and 40 K.

The γ-γ cascade of 329,487 keV populated from the β^- decay of ^{140}La was used for the measurement of magnetic interaction at the 2083 keV intermediate state (4^+ spin) of ^{140}Ce. TDPAC spectra were recorded at several temperatures, in the range from 8 to 295 K, using a standard setup with four BaF_2 detectors arranged in a planar 90°–180° geometry. Twelve delayed coincidence spectra are generated simultaneously with a time resolution of 600 ps. For low-temperature measurements the sample was attached to the cold finger of a closed-cycle-helium refrigerator with temperature control (better than 0.1 K).

3. Results and discussion

Figure 1 shows the results for the magnetization measurements as a function of the applied magnetic field at 5 and 40 K. The magnetization at low field and low temperature shows the characteristic linear behavior expected from a antiferromagnetic system, and the results at 40 K exhibit the typical behavior of a paramagnetic phase. However, it was observed that the magnetization does not saturate at high magnetic fields (see inset a). The antiferromagnetic phase is better characterized by the measurement of the magnetization as a function of temperature for 1.0 kOe (inset b), where the magnetization increases when the temperature increases. This behavior is a characteristic of an antiferromagnetic

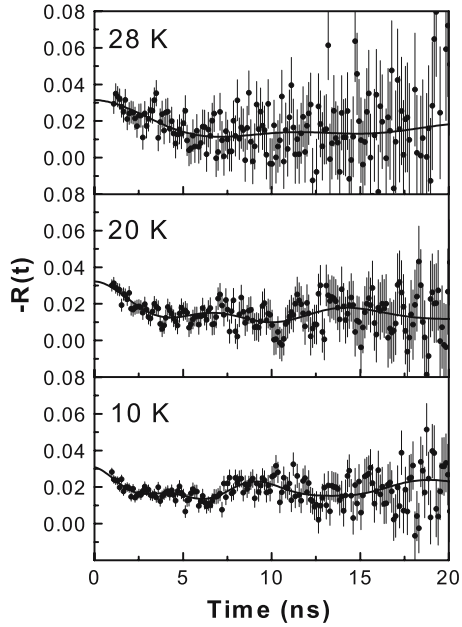

Figure 2. Perturbation functions $R(t)$ for ^{140}Ce in Tb_3In_5 measured at indicated temperature.

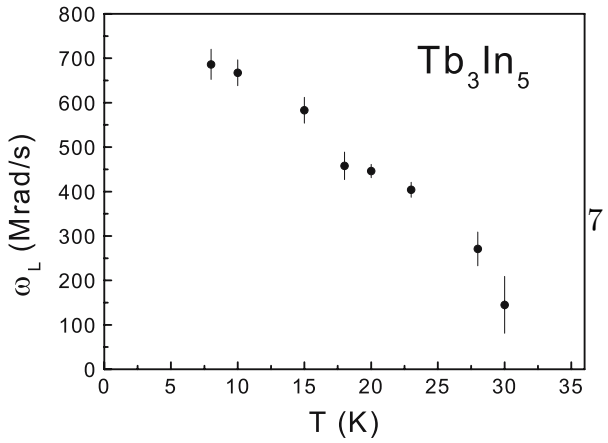

Figure 3. The temperature dependence of fitted Larmor frequency for ^{140}Ce at Tb site in Tb_3In_5.

phase. The Néel temperature (T_N ~27 K) was determined at the peak of the magnetization curve, above of which the magnetization decreases, as expected for a paramagnetic phase.

The spin rotation spectra $R(t)$ is well described by a superposition of two fractions or sites with pure temperature dependent magnetic interactions. The proportion between these fractions is approximately 1:3, as expected for the two Tb sites in the orthorhombic structure of Tb_3In_5. The major fraction (~65%),with

distribution $\delta \sim 13\%$, shows higher frequencies than the minor fraction ($\delta \sim 6\%$). Some of the TD-PAC spectra measured with ^{140}Ce probe nuclei are shown in Figure 2. The solid curves are the least squares fit of the experimental data to the appropriate function.

The temperature dependence of the Larmor frequency ω_L corresponding to the major fraction is plotted in Figure 3. The observed magnetic interaction corresponds to the antiferromagnetic ordering of the Tb moments. If the points for 18 and 20 K are not taken into account, one could consider that the data follow a Brillouin function. But, if all data are considered true, they indicate that instead of approaching a saturation value around 500 Mrad/s, ω_L values increases at lower temperatures below around 18 K, and point to a saturation value of $\sim$720 K. This behavior was already observed in $CeMn_2Ge_2$ compound [10] and was attributed to the spin polarization of Ce ions of the probe nuclei at low temperatures induced by the magnetic field from rare earth moments.

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