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MAGNETIC ANISOTROPY OF TERNARY (Tb\textsubscript{x}Gd\textsubscript{1-x})Al\textsubscript{2} COMPOUNDS

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Abstract. - Magnetization measurements, parallel and perpendicular to the magnetic field have been performed on crystals of Tb\textsubscript{x}Gd\textsubscript{1-x}Al\textsubscript{2}. We determine the crystal field parameters by fitting the field and temperature dependences of the magnetization angle with [001]. The \(B_{4}\) and \(B_{6}\) CEF parameters are almost \(x\) independent, and \(B_{4} \approx 5 \times 10^{-4} \text{ K/ion}\) for TbAl\textsubscript{2}.

1. Introduction

The Tb\textsubscript{x}Gd\textsubscript{1-x}Al\textsubscript{2} intermetallics have the cubic Laves phase structure and order ferromagnetically with Curie temperatures between 100 K and 145 K \cite{[1]}. Anisotropy is expected to issue from the Tb\textsuperscript{3+} ion, albeit possible anisotropic exchange where the Gd 5d electrons could play a role. Moreover the easy direction is [111] \cite{[1]}. In previous works \cite{[1], [2]} we demonstrated that anisotropy was single-ion, and that the same cubic crystal field (CEF) parameters were able to explain the magnetic properties of such compounds.

This investigation aims to determine also the CEF interaction, in special to study in a detailed way the evolution of the CEF parameters. For that we have rotated the Tb\textsuperscript{3+} (\(\mu_{\text{Tb}}\)) and Gd\textsuperscript{3+} (\(\mu_{\text{Gd}}\)) magnetic moments, within the plane (110), by use of a magnetic field \((H = 2.05 \text{ T})\), and measuring the parallel \((M_{\parallel})\) and perpendicular \((M_{\perp})\) magnetization components.

This enables us to determine the modulus, \([M]\), and angle of rotation \(\theta\) of \(M\) with respect to [001]. As a by product we determined the anisotropy constant, \(K_{1}\), although due to the strong anisotropies only the value of \(K_{1}\) for low \(x(=0.2)\) was found reliable. We should notice that the anisotropy torque is \(L_{k} = M_{\perp}H\), and therefore the measurements of \(M_{\perp}\) is a good tool in determining magnetic anisotropy.

2. Outline of theoretical model

We will outline a mean-field two-dimensional model \cite{[3]}, to explain the rotation of Tb\textsuperscript{3+} and Gd\textsuperscript{3+} moments, and which takes into account a non-collinearity between such moments. Although the Gd\textsuperscript{3+} ions do not feel the CEF, they interact via exchange with Tb\textsuperscript{3+} moments, and therefore we shall have a canting angle, \(\varepsilon\), between both. We denote by \(\phi\) the angle between \(\mu_{\text{Gd}}\) and [001]. Briefly speaking those compounds are well described by a Tb\textsuperscript{3+} ionic Hamiltonian which reads,

\[ H_{\text{TB}} = H_{\text{CEF}} + H_{a} + H_{z} \quad (1) \]

where the CEF hamiltonian is,

\[ H_{\text{CEF}} = B_{4} (O_{4} - 5O_{0}^{4}) + B_{6} (O_{6}^{0} + 21O_{4}^{0}) \quad (2) \]

where \(O_{n}\) are Stevens operators, the Hamiltonian being referred to axis rotated \(\pi / 4\) around [001]. The exchange Hamiltonian is assumed to be

\[ H_{\text{ex}} = (1 - x) H_{\text{ex}}^{\text{TB,Gd}} + x H_{\text{ex}}^{\text{TB,Tb}} \quad (3) \]

where \(H_{\text{ex}}^{\text{TB,Gd}}\) describes the interaction between Tb\textsuperscript{3+} and Gd\textsuperscript{3+} ions, and \(H_{\text{ex}}^{\text{TB,Tb}}\) the one between Tb\textsuperscript{3+} ions. \(H_{z}\) is the Zeeman term. A description of Hamiltonians \((3)\) may be found in \cite{[3]}, and include as variable \(\phi + \varepsilon\). The free energy is evaluated as \(F = -K_{b}T \ln Z\) where the partition function \(Z\) is evaluated with the eigenvalues of \((1)\). Also taken into account is the energy of the Gd\textsuperscript{3+} sublattice, which provides with a relation between \(\phi\) and \(\varepsilon\). The procedure now is to minimize \(F\) with values of \(B_{4}\), \(B_{6}\) that provide the equilibrium ones for \(\phi\), \(\varepsilon\).

3. Experiment

Magnetization measurements, down to 4.2 K, were performed for \(x = 1.0, 0.8\) and 0.2, with applied fields, \(H\), up to 2.05 T. Magnetizations \(M_{\parallel}\) and \(M_{\perp}\) to \(H\) have been determined, with a magnetometer for such purpose. Isotherms of \(M_{\parallel}\) and \(M_{\perp}\) against the angle of \(H\) with [001], \(\psi\), are given in figure 1. \(M_{\parallel}\) is maximum when \(H\parallel[111]\) and minimum for \(H\parallel[001]\). On the other hand \(M_{\perp} = 0\) when \(H\) crosses high symmetry directions \((L_{k} = 0)\). This behaviour can be explained considering that the crossing through the hard directions occurs with decomposition in two magnetic domains, giving null \(M_{\perp}\), but non-null \(M_{\parallel}\). Noteworthy is that the magnetization modulus \((|M| = [M_{\parallel}^{2} + M_{\perp}^{2}]^{1/2})\) remains constant within the rotation. From figure 1 isothersms we can determine the angle \(\theta\) as \(\theta = \tan^{-1} (M_{\perp} / M_{\parallel}) + \psi\). The canting angle \(\varepsilon\) was difficult to determine, being smaller than 2°.
From Fourier analysis of \( M_4 \) isotherms (Fig. 1) we determine \( K_1 \). Due to the large anisotropies, the torque \( L_k \) did not saturate, except for \( x = 0.2 \). A scaling with reduced magnetization, \( m \), of the form \( m^a \) with \( a = 11.6 \) was obtained, which deviates from \( a = 10 \). A plot of \( K_1 \) vs. \( x \) shows good linearity only with data below \( x = 0.2 \) [4].

4. Results and discussion

In figure 2 we show the field and temperature dependences of angle \( \theta \) of \( M \) with [001], with \( \mathbf{H} \) applied along [001]. \( \theta \) was obtained as \( \theta = \cos^{-1} \left( M_{||[001]} / M_{||[111]} \right) \), because \( |M| = M_{||[111]} \). As expected the slope \( \partial \theta / \partial H \) decreases with temperature. In the same figure 2 we plot the theoretical fits using the model of paragraph 2. The fits are good considering the ranges of fields and temperatures spanned. The obtained CEF parameters, \( B_4 \) and \( B_6 \), are: for \( x = 1 \), \( B_4 = 5 \times 10^{-4} \) and \( B_6 = -5.3 \times 10^{-6} \); for \( x = 0.8 \), \( B_4 = 4.2 \times 10^{-4} \) and \( B_6 = -5.4 \times 10^{-6} \); for \( x = 0.2 \), \( B_4 = 3.8 \times 10^{-4} \) and \( B_6 = -2.2 \times 10^{-6} \) (all values in K/\( \text{ion} \)). As we can observe, there is a weak variation across the series. Therefore the constancy of the CEF parameters points out to single-ion origin for magnetic anisotropy, confirming previous results [1, 2]. A comparison with the usually quoted values for \( \text{TbAl}_2 \) [5, 6] i.e. \( B_4 = 3.5 \times 10^{-3} \text{K/\( \text{ion} \)}, B_6 = +2.9 \times 10^{-6} \text{K/\( \text{ion} \)}, \) shows that our \( B_4 \) values are one order of magnitude smaller, and \( B_6 \) is of the same order but of opposite sign. We should mention that fitting the angle \( \theta \) within a so large ranges of field and temperature is a rather strict test.

Another result is the constancy of \( |M| \) with rotation in plane (110); this is an indication of absence of magnetization anisotropy, and of quenching of CEF upon \( \mu_{\text{Tb}} \), which preserves its free ionic moment \( (\approx 9 \mu_B) \).

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