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Magnetic and electronic properties of nanocrystalline $Gd_3Fe_5O_{12}$ garnet

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ABSTRACT

The $Gd_3Fe_5O_{12}$ nanocrystalline Gadolinium Iron Garnet (GdIG) obtained from a sintered block was milled in a high energy ball mill. We measured the magnetization at 5 K under applied fields up to 12 T. We report here our study of approach to saturation magnetization. The results have been interpreted within the framework of random anisotropy model. From an analysis of the approach to saturation magnetization some fundamental parameters have been extracted. We have determined the anisotropy field H_r and the local magnetic anisotropy constant K_L. In addition, first-principles spin-density functional calculations, using the Full potential Linear Augmented Plane Waves (FLAPW) method are performed to investigate electronic and magnetic structures. All computed parameters are discussed and compared to available experimental data.

1. Introduction

Nanocrystalline magnetic materials are being currently studied in several laboratories. One of them is the ferromagnetic oxides, such as the spinels and garnets. These oxide materials are attractive for certain applications in the microwave region hence much attention is being given to them. There are essentially two types of nanomaterials that are being studied, namely, the nanopowder obtained by power ball milling of the bulk sintered materials and thin films prepared either by sputtering or by pulsed laser deposition techniques. There is a plethora of publications on such materials dealing with the preparation and study of their magnetic properties [1–3]. The properties of the nanomaterials are found to be different from their bulk counterpart. It is generally observed that for nanomaterials both magnetization and the Curie temperature show a decrease. However in some cases novel behaviour is found. For instance, reduction in grain size leads also to changes in the cation distribution, magnetization and Néel temperature [4,5]. As regards garnets the results are somewhat surprising. Both $Y_3Fe_5O_{12}$ Yttrium iron (YIG) and $Gd_3Fe_5O_{12}$ Gadolinium iron garnets (GdIG) when ball milled to produce nanocrystalline material, below a certain critical size, showed a phase decomposition resulting in the formation of orthoferrite [6]. We have reported in detail our study on nanocrystalline GdIG and also discussed the magnetic properties [7]. Besides such changes, there is a common behaviour in most of the nanospinels and garnets both as thin films and nanopowders that is the difficulty to attain magnetic saturation. In their bulk state these materials which are cubic could be saturated with moderate applied fields of the order of 0.5 T, whereas in their nanostate, they do not show saturation even at 2 T. This has been reported by many authors working on nanograin ferrites who invoke several models to explain the small but definite positive slope in the M–H curve near the saturation point [8,9]. This slope is termed as high field susceptibility and some authors have analysed this to some extent.

In this paper, the anisotropy in nanocrystalline $Gd_3Fe_5O_{12}$ garnet is calculated and discussed. Fundamental parameters of such nanocrystalline are estimated as well. In addition, electronic and magnetic structures calculations, using the FLAPW method, are reported.

2. Experimental

Gadolinium iron garnet obtained from a sintered block was milled in air in a Fritsch P7 high energy ball milling using zirconia balls and vial. The ball to powder weight ratio was 10:1. The magnetic measurements were performed using a quantum design SQUID magnetometer for fields up to12 T at 5 K. X-ray diffraction (XRD) measurements were carried out using a Fe target. The average grain size D was calculated from the XRD measurements using the Scherrer formula.

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3. Electronic structure calculations

We used the FLAPW method [10] which performs the density functional theory (DFT) calculations using the General Gradient Approximation (GGA) where the Kohn–Sham equation and energy functional are evaluated self-consistently. For this method, the space is divided into the interstitial region and the non-overlapping muffin tin spheres centred on the atomic site. The employed basis function inside each atomic sphere is a linear expansion of the radial solution of a spherically potential multiplied by spherical harmonics. In the interstitial region the wave function is taken as an expansion of plane waves and no shape approximation for the potential is introduced in this region; which is consistent with the full potential method convenient to nonmetallic compounds. The core electrons are described by atomic wave functions which are solved fully relativistically using the current spherical function base and the valence electrons are treated with the spin polarized potential in our case.

The crystallographic structure as reported in Ref. [11] is defined in the space group *Ia*-3*d* (#230) with 4 independent atoms: gadolinium, oxygen and the two iron atoms successively at 24*c*, 96*h*, 16*a* (octahedral) and 24*d* (tetrahedral) sites. The GdIG material is considered to be in the collinear magnetic state where the magnetic moments of octahedral and tetrahedral sites are opposite. The atomic muffin-tin (MT) spheres, supposed not to overlap with each other, are taken as 2.26, 1.8, and 1.6 a.u for Gd, Fe and O, respectively. The gap energy, which defines the separation of the valence and core state, was chosen equal to -6.0 Ry. The largest reciprocal vector G in the charge Fourier expansion, G_{max}, was equal to 14 and the cut-off energy corresponding to the product of the muffin-tin radius and the maximum reciprocal space vector, $R_{MT} \times k_{max}$, was equal to 7. Inside the atomics spheres, the potential and charge density are expanded in crystal harmonics up to $l_{max}=10$. Calculations are performed with 20 inequivalent k-points in the irreducible Brillouin. Such value is sufficiently large to ensure the spin–spin moment. The convergence criterion was chosen to be the total energy and set at 10^{-4} eV.

4. Results and discussion

Fig. 1 shows the XRD pattern of the as-prepared and 10 h milled samples. The 10 h milled sample shows the decomposition of the garnet phase to gadolinium orthoferrite and Gd_2O_3. The average grain size for the as-prepared and 10 h milled samples is 78 and 33 nm, respectively. Fig. 2 shows the field dependence of magnetization at 5 K for the as-prepared and 10 h milled samples. The magnetization at 5 K decreases with milling, due to increase in the

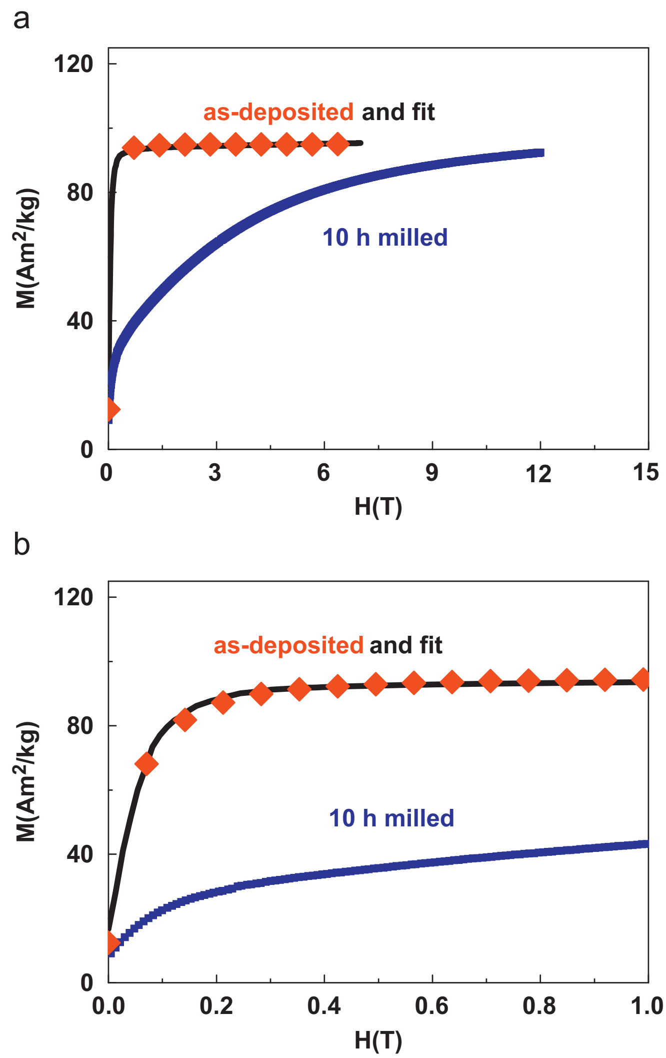

Fig. 2. Experimental curve of $M(H)$ for the as deposited and 10 h milled GdIG samples at 5 K and the fit by the model used. (a) H varies from 0 to 12 T and (b) H varies from 0 to 1 T.

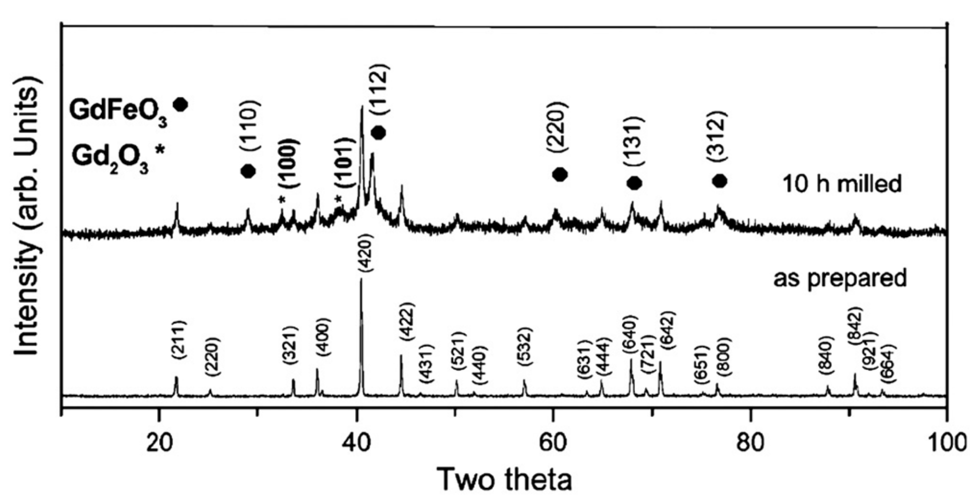

Fig. 1. XRD spectrum for the as deposited and 10 h milled GdIG samples. The symbols indicate the positions of $GdFeO_3$ and Gd_2O_3.