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Pulsed Laser Deposition of permanent magnetic $\text{Nd}_2\text{Fe}_{14}\text{B}$ thin films

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Abstract

Pulsed Laser Deposition (PLD) is applied to deposit thin (thickness typically 100 nm) films of $\text{Nd}_2\text{Fe}_{14}\text{B}$. It is shown that films can be grown which have the desired composition and phase. $\text{Nd}_2\text{Fe}_{14}\text{B}$ grows with the *c*-axis along the film normal on $\langle 110 \rangle$ Al_2O_3 single crystal substrates covered with a Ta layer. These films are found to have a strong magnetic anisotropy along this axis.

1. Introduction

The rare-earth transition metal intermetallic compounds are a class of materials containing the strongest permanent magnets known to date [1]. More specifically, materials of the $\text{Nd}_2\text{Fe}_{14}\text{B}$ family are widely used as permanent magnets with a high energy product. Although a considerable amount of research has been performed on the bulk properties of such materials, attention to thin films, and in particular films with well-controlled composition and crystallography, has been rather modest [2–4]. Such films would be interesting both for direct application (as a recording medium or in micromechanical applications), and as well-controlled model sys-

tems which would allow the study of coercivity mechanisms determining the magnetic performance of bulk materials. This limited progress can be attributed to the complex crystallographic and stoichiometric nature of these alloys, which hampers successful preparation of high quality films by conventional thin film deposition methods.

In this work, we report on the results of initial experiments exploring the possibility of preparation of high quality $\text{Nd}_2\text{Fe}_{14}\text{B}$ films by Pulsed Laser Deposition (PLD). Both structural and magnetic properties have been investigated.

2. Experimental

All films have been deposited in a load-locked UHV-PLD system which has been described earlier [5,6]. Up to 4 targets are irradiated by 308 nm/27 ns

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laser pulses with laser fluences up to 10 J cm^{-2} . A circular diaphragm was introduced in the laser beam in order to increase beam homogeneity. Furthermore, the laser beam was scanned over the target surface in order to obtain a target use which is as homogeneous as possible. In order to minimize the amount of particulates on the films, a mechanical velocity filter ('chopper') was used which operated at 100 Hz rotation speed. Background pressures during deposition were typically of the order of 3×10^{-8} Torr. The results as presented here have been obtained on films (thickness typically 100 nm) grown on $\langle 110 \rangle$ Al_2O_3 single crystal substrates which can be heated by a filament oven up to 700°C .

Film composition was measured using Rutherford Backscattering Spectrometry (RBS), and Electron Probe MicroAnalysis (EPMA). Structural characterisation on the films was performed using X-Ray diffraction (θ - 2θ scans).

A Vibrating Sample Magnetometer (VSM) was used to measure magnetization loops ($-2150 \text{ kA/m} < H < 2150 \text{ kA/m}$) both along the film normal and

in the film plane. Furthermore, magnetization torque measurements have been used to determine the anisotropy direction of the film.

3. Results and discussion

The film composition is discussed first. The deposition was not exactly stoichiometric. All films were found to display a Nd loss with respect to the target, which was more pronounced for higher laser fluences ($> 5 \text{ J cm}^{-2}$). Furthermore, films grown at elevated temperatures (typically 650°C and above) showed a decreasing B-content with increasing temperature, most probably due to evaporation from the growing film. To illustrate the extent of these deviations, let us note that a film ablated from a slightly B and Nd enriched commercial ingot target (Rhône-Poulenc; target composition: 13.5 at% Nd, 78.9 at% Fe, 7.6 at% B ($\text{Nd}_{1.77}\text{Fe}_{10.4}\text{B}_1$)) at a fluence of 4 J cm^{-2} on a substrate with a temperature of 630°C has a composition of: 11 at% Nd, 83.7 at% Fe, 5.3%

Fig. 1. θ - 2θ scan (Cu $\text{K}\alpha$ radiation) of a 100 nm thick $\text{Nd}_2\text{Fe}_{14}\text{B}$ film grown on a Ta-buffered Al_2O_3 substrate.

B ($\text{Nd}_{2.1}\text{Fe}_{15.8}\text{B}_1$). It is thus found that films can be obtained which have relative concentrations within a few % equal to the ideal 2:14:1 composition.

Deposition of the $\text{Nd}_2\text{Fe}_{14}\text{B}$ directly on the Al_2O_3 substrate at high temperatures was found to lead to a reaction between the Nd in the film and the oxygen in the substrate. Therefore, a Ta film (thickness typically 15 nm) was deposited in between the $\text{Nd}_2\text{Fe}_{14}\text{B}$ film and the substrate, as well as on top of the film, to prevent oxidation. Due to its higher ablation threshold, the Ta layer requires a somewhat higher laser fluence (8 J cm^{-2}).

Having established process conditions which lead to an acceptable stoichiometry, we proceed by characterizing the structure of the films obtained under these conditions. Fig. 1 shows a typical θ - 2θ scan ($\text{Cu K}\alpha$ radiation) of a 100 nm thick film. Apart from the two intense substrate reflections, one finds 5 intense reflections. These can be labeled as $\text{Nd}_2\text{Fe}_{14}\text{B}$ (00 l) reflections ((004); $2\theta = 29.34^\circ$, (008); $2\theta = 60.89^\circ$ and (0012); $2\theta = 99.10^\circ$), as a Ta (200) reflection (at $2\theta = 33.7^\circ$) and a reflection which can be attributed to either the $\text{Nd}_2\text{Fe}_{14}\text{B}$ (006) line ($2\theta = 44.65^\circ$) or the Fe(110) line ($2\theta = 44.75^\circ$). It is thus seen that the film consists mainly of the $\text{Nd}_2\text{Fe}_{14}\text{B}$ -phase, although it cannot be excluded a priori that some fraction of the film would be in the

alpha Fe phase. In fact, films with low Nd content are found to display a relatively intense peak at $2\theta = 44.7^\circ$ indicating the presence of alpha Fe in these films, as expected from the Nd-Fe-B-ternary phase diagram [1]. However, as the composition of optimized films is close to the ideal $\text{Nd}_2\text{Fe}_{14}\text{B}$ composition, and no evidence for other Nd-containing phases was found, it is expected that a major fraction of these optimized films are in the $\text{Nd}_2\text{Fe}_{14}\text{B}$ phase.

As the θ - 2θ scan displays only $\text{Nd}_2\text{Fe}_{14}\text{B}$ (00 l) lines, the films are shown to be textured with the c -axis along the film normal. This is in agreement with earlier work [3] showing the occurrence of a similar texture in films sputter-deposited on polycrystalline Ta-foils.

Fig. 2 shows typical magnetic measurements. Both in-plane and perpendicular magnetization loops are shown. It can be seen that the perpendicular loop saturates for field values around 1300 kA/m at a magnetisation around 1.3 T. The loop clearly shows hard magnetic behaviour with a coercivity around 400 kA/m, and a remanent magnetisation of 0.64 T. These values are comparable to the values which have been found earlier in sputtered films [2–4] with c -axis texture. However, they are much lower than the values as found in bulk magnets i.e. coercivities above 1000 kA/m and remanent magnetizations of

Fig. 2. In plane and perpendicular magnetisation loops of a 100 nm thick $\text{Nd}_2\text{Fe}_{14}\text{B}$ film.

1.3 T (Philips magnetic products Res421). Apparently, domain wall processes, which determine these quantities, differ in the present films from those in sintered bulk magnets. The in-plane loop cannot be saturated within the present field range. It displays a considerably lower coercivity and remanence than the perpendicular loop. These loops suggest the presence of a considerable uniaxial anisotropy (anisotropy fields of several Tesla, beyond the measuring range of our equipment) along the normal of these films, as can be understood from their *c*-axis orientation, and the well-known fact that the anisotropy in $\text{Nd}_2\text{Fe}_{14}\text{B}$ is along the *c*-axis [1]. Torque measurements show indeed the presence of such an anisotropy along axes which are, although varying from film to film, always within 10° from the film normal. It has been observed earlier [4] in sputtered films that a close correlation exists between the degree of *c*-axis orientation and the occurrence of a magnetic behaviour similar to the one shown in Fig. 2.

4. Conclusions

It is found that PLD can be used to produce films of $\text{Nd}_2\text{Fe}_{14}\text{B}$ on Ta-buffered Al_2O_3 with the desired 2:14:1 composition and a growth with the *c*-axis

along the film normal. Such films are found to display a strong perpendicular anisotropy which results in coercivities around 400 kA/m along the film normal.

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