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Magnetic and structural properties of high-coercivity $Pr_2Fe_{14}BPr$ cosputtered films

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Magnetic and structural properties of high-coercivity $Pr_2Fe_{14}B$:Pr cosputtered films

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The results of structural and magnetic measurements on a series of films produced by cosputtering $Pr_{21}Fe_{72}B_7$ and Pr are reported. The materials were deposited serially, 10–200 Å at a time, onto 700 °C Ta substrates attached to a computer-controlled stepping motor. The structure of the resulting films was examined by large- and small-angle x-ray diffraction, scanning electron microscopy, and by microprobe analysis. Magnetic measurements were made at temperatures from 4.2 to 300 K, in fields up to 80 kOe in a vibrating sample magnetometer. The films appear to consist of 1-μm grains of $Pr_2Fe_{14}B$ with their c axes oriented perpendicular to the film plane, surrounded by a Pr-rich phase. The maximum room-temperature coercivity discovered so far is almost 20 kOe, which is close to the values reported for bulk $Pr_2Fe_{14}B$ magnets.

I. INTRODUCTION

During the last few years we have studied the effects of sputtering conditions on the microstructure and magnetic properties of the $Nd_2Fe_{14}B$ phase in films and multilayers sputtered from a cobalt-doped 2:14:1 target.[1-3] In all of the work with NFB films and multilayers, the spin reorientation at about 150 K greatly complicates the interpretation of the magnetization data. For this reason we recently began studying films and multilayers based on $Pr_2Fe_{14}B$ (PFB), which does not exhibit a spin reorientation.[4] In particular, in this paper we report our results on several films of the form PFB:Pr, emphasizing structural aspects and the field and temperature dependence of the magnetic properties. Pr was chosen as the interlayer because it has a large single-ion anisotropy and should couple ferromagnetically with the magnetic elements of the PFB layer. Thus it was thought to have potential for enhancing the hard magnetic properties of the PFB phase.

II. EXPERIMENTAL METHODS

The samples were prepared on Ta substrates in a multiple-gun sputtering system with a temperature-controlled substrate holder.[5] The holder was precisely positioned over the appropriate dc sputtering gun by a computer-controlled stepping motor. A homemade $Pr_{21}Fe_{72}B_7$ target, produced by arc melting the appropriate amounts of Pr (99.9% pure), Fe (99.9% pure), and B (99.5% pure), was placed in one gun and a Pr (99.9% pure) target was placed in the other. The sputtering rate was about 7 Å/s for the $Pr_{21}Fe_{72}B_7$ and about 8 Å/s for the Pr. The substrate temperature was held constant at temperatures between 500 and 800 °C during sputtering. The first and last layer deposited was PFB, and the number of layers PFB deposited was adjusted so that the

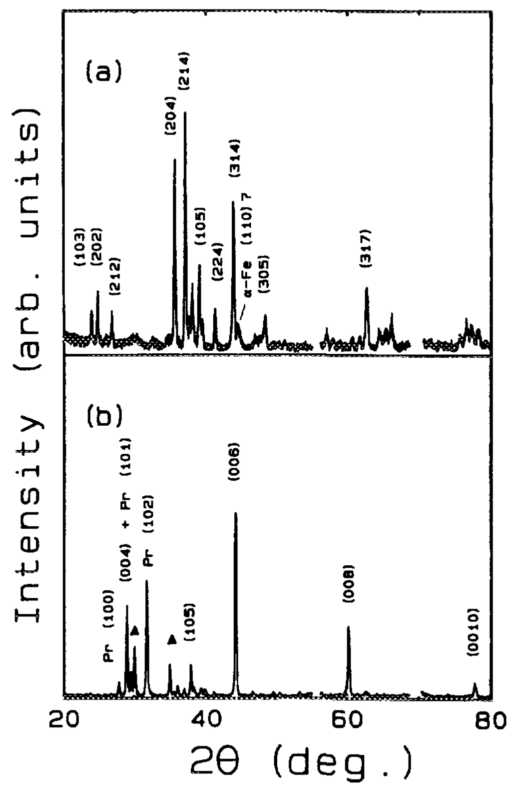

FIG. 1. (a) Large-angle x-ray diffraction pattern for an undoped PFB film. The major lines from the 2:14:1 phase are labeled. This figure indicates the possibility of α-Fe in the film. The lines associated with the fcc PrO phase are too small to readily discern on this scale. (b) X-ray diffraction pattern for a $Pr_2Fe_{14}B$(200 Å):Pr(40 Å) film. The lines from the 2:14:1 phase are labeled along with the major lines from α-Pr. The lines marked with the triangles are from the fcc PrO phase, and the remaining unlabeled lines are from these three phases. The high intensity of the 00l relative to the other lines from the 2:14:1 phase, e.g., the (105) line, indicate that the c axes of the grains are perpendicular to the film plane. The Ta substrate lines have been removed for clarity (gaps).

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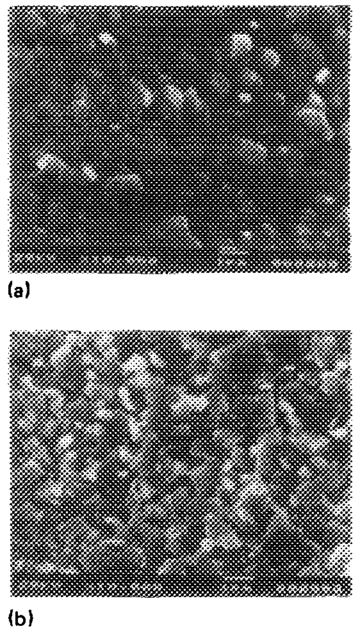

FIG. 2. SEM picture of the (a) unetched, and (b) etched surface of a $Pr_2Fe_{14}B$(200 Å):Pr(40 Å) film. Microanalysis indicates that all of the grains are nearly pure 2:14:1 phase.

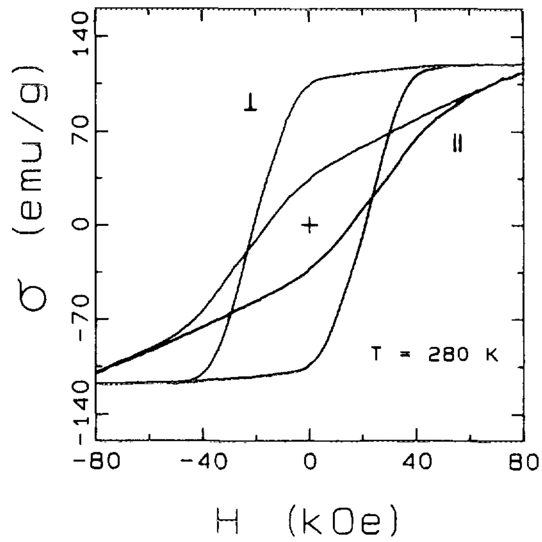

FIG. 4. Typical hysteresis loops, for PFB(200 Å):Pr(44 Å) film, measured with the field applied parallel to the plane (∥) and perpendicular to the plane (⊥).

total PFB thickness was about 1 μm; e.g., for a PFB(200 Å):Pr(10 Å) film, 50 layers of PFB and 49 layers of Pr were deposited.

The structure of the films was determined by small- and large-angle x-ray diffraction with a Rigaku diffractometer and by scanning electron microscopy (SEM) with a JEOL JSM-T330 microscope. The composition of several samples was verified by a Kevex microanalyzer with a quantum detector attached to the SEM. Some of the SEM samples were etched for several seconds in a bath containing 2 ml nitric acid, 100 ml methanol, and 5 ml water (this mixture is also known as 2% nital) to reveal grain boundaries.

The magnetization measurements were made at temperatures from 4.2 to 300 K and fields up to 80 kOe in a vibrating sample magnetometer (VSM) using a specially designed heated probe with low moment.

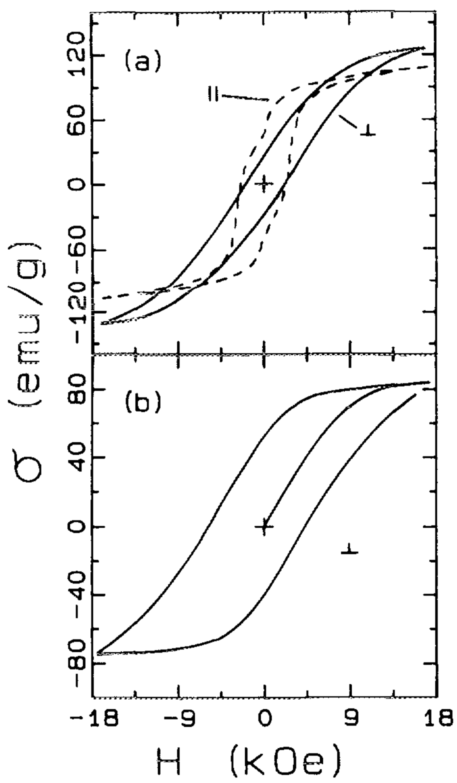

FIG. 3. Room-temperature hysteresis loops with a maximum field of 17.1 kOe for (a) an undoped PFB film, and (b) a virgin PFB(200 Å):Pr(10 Å) film.

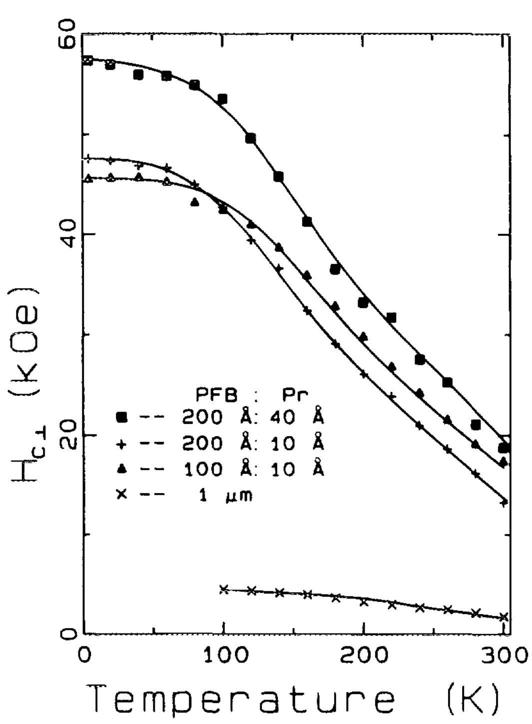

FIG. 5. Summary of coercivity vs temperature data with the field applied perpendicular to the film plane.

III. RESULTS AND DISCUSSION

The first films we made were sputtered directly from the $Pr_{21}Fe_{72}B_7$ target with no additional Pr added. Large-angle x-ray diffraction patterns [Fig. 1(a)] for these films show peaks that can be associated with the tetragonal 2:14:1 phase, perhaps α-Fe, and a small amount of an fcc phase that is tentatively identified as PrO or boron-substituted PrO.[2]

We also attempted to make PFB:Pr multilayers, varying the PFB thickness from 50 to 200 Å and the Pr thickness from 10 to 40 Å. Figure 1(b) is a large-angle x-ray diffraction pattern for a nominally PFB(200 Å):Pr(40 Å) film. The 00l lines are more intense than the other lines for the 2:14:1 phase, indicating that the c axes of the 2:14:1 phase are perpendicular to the film plane. Preliminary rocking-curve measurements about the (006) line give a mosaic spread of about 5°. This film also contains some excess Pr and a small amount of PrO. However, x-ray diffraction did not show any low-angle peaks that would confirm compositional modulation, at least for Pr thicknesses between 10 and 40 Å.

SEM studies of the undoped films reveal a remarkably smooth microstructure with few grain boundaries visible on any scale. Microanalysis of the unetched surface of these films indicates that the ratio Fe to Pr is close to 8:1, suggesting that there is a second Fe-rich phase (perhaps α-Fe) in the films.

Figure 2 shows an SEM picture of the surface of an (a) unetched, and (b) etched PFB(200 Å):Pr(40 Å) film; it clearly shows a predominance of 1-μm square grains, with a distribution of somewhat smaller grain sizes. Microanalysis of the etched surface reveals an Fe to Pr ratio close to 7:1, as expected for pure $Pr_2Fe_{14}B$. The unetched surface has a higher concentration of Pr, suggesting that the films consist of large 2:14:1 particles surrounded by a thin layer of Pr or a Pr-rich phase.

Figure 3(a) shows typical room-temperature hysteresis loops for an undoped PFB(200 Å):Pr(10 Å) film, with a maximum applied field of 17.1 kOe. The undoped films exhibit perpendicular anisotropy, and the loop measured with the field applied parallel to the film plane is constricted in the low-field region. This constriction is consistent with the existence of a soft magnetic phase in the film. The maximum room-temperature coercivity for the undoped films is about 5 kOe.

A virgin magnetization curve for a nominally PFB(200 Å):Pr(10 Å) film measured at room temperature is shown in Fig. 3(b). The initial susceptibility is about 0.4 (corrected for the $4\pi\langle M\rangle$ demagnetizing field, assuming the density calculated from the ideal layer thicknesses and densities for Pr and $Pr_2Fe_{14}B$). Notice that the minor loop is offset from the origin along the magnetization axis. These properties are typical of all Pr-doped films studied to date.

Typical in-plane and perpendicular hysteresis loops for the Pr-doped films, measured in a maximum field of 80 kOe, are shown in Fig. 4. This figure illustrates the large perpendicular anisotropies and coercivities found in these films.

Figure 5 gives a summary of the coercivity versus temperature with the field applied perpendicular to the film plane for all of the PFB:Pr samples measured to date. A coercivity versus temperature curve, measured down to $T = 100$ K, for an undoped PFB film is included for comparison. It should be noted that the maximum room-temperature coercivity for the Pr-doped films is almost 20 kOe, close to the values reported for sintered bulk magnets of this material.[6,7]

IV. CONCLUSIONS

Particulate $Pr_2Fe_{14}B$ films with large coercivities can be produced by cosputtering from a $Pr_{21}Fe_{72}B_7$ target and a pure Pr target. These films consist of 1-μm grains of $Pr_2Fe_{14}B$, oriented with their c axes perpendicular to the film plane, perhaps surrounded by a thin layer of Pr or Pr-rich phase.

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