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Orientation of hyperfine magnetic fields of α-iron films produced by laser deposition

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Abstract Iron films were produced by pulsed laser deposition (PLD) of iron in Ar gas and Mössbauer spectra of these films were obtained at room temperature. The orientation of the hyperfine magnetic field was found to vary depending on the pressure of the Ar gas. Iron films produced at low Ar pressures exhibited magnetic fields parallel to the substrate surface. The magnetic field became increasingly perpendicular to the substrate with increasing Ar pressure. Collisions with Ar gas molecules reduced the translational energies of laser-evaporated iron atoms and thus the orientation of crystals formed on the substrate varied depending on the Ar pressure.

Keywords Iron films · Pulsed laser deposition · Magnetic field orientation · Ar ambient gas

1 Introduction

The magnetic properties of thin films have been extensively studied, especially the magnetic field orientation dependence on the thin film shape. Mössbauer spectroscopy is a very useful technique for investigating magnetic properties and for determining the orientation of magnetic fields of iron-based materials. The intensity ratio ($I_{2,5}/I_{3,4}$) of sextet absorption in Mössbauer spectra varies depending on the angle between a γ-ray and the nuclear spin.

$$\frac{I_{2,5}}{I_{3,4}} = \frac{4\sin^2\theta}{1+\cos^2\theta}$$

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We have previously reported that the orientation of magnetic fields of iron films can be controlled by employing different deposition methods. Pulsed laser deposition (PLD) of Fe in a vacuum produces an iron film with a magnetic field parallel to the substrate surface [1], while deposition of Fe vaporized by resistive heating produces a film with a magnetic field perpendicular to the substrate surface [2]. Deposition of Fe atoms or ions vaporized by an arc plasma gun produces a film with a magnetic field aligned with the grooves on the substrate [3]. The magnetic field orientation is strongly influenced by the Fe vapor density and the translational energy; Fe vapors produced by laser evaporation have energies higher than 100 eV, Fe atoms vaporized by resistive heating have energies of several eV, and Fe atoms vaporized by an arc plasma gun have energies lower than 100 eV. We have previously reported PLD in ambient gases (O_2, N_2, and C_2H_2) to produce films of iron-based compounds (oxides, nitrides, and carbides) [4, 5]. In this study, we produced films on Al substrates by PLD of Fe in an Ar atmosphere while controlling the energy of the Fe atoms.

2 Experimental

A block of Fe metal in a vessel filled with Ar gas was laser evaporated using a Nd:YAG laser (NewWave, Tempest 10; wavelength: 523 nm; pulse energy: 85 mJ; repetition rate: 10 Hz). The Ar pressure was maintained at the target pressure between 10^{-7} and 20 Torr using a pressure controller. An Al substrate (thickness: 40 μm) was placed 10 mm from the Fe target. The film thickness was estimated by measuring the difference between the masses of the substrate before and after PLD. Mössbauer spectra of the Fe films were obtained at room temperature and their surface morphologies were observed by scanning electron microscopy (SEM).

3 Results and discussion

Fe films were produced by PLD on Al substrates for various Ar pressures in the range 10^{-7} to 10 Torr and their Mössbauer spectra were measured at room temperature (Fig. 1). The films were less than 1 μm thick. The film produced at the lowest Ar pressure (10^{-7} Torr) exhibited sextet absorption with an area intensity ratio $I_{2,5}/I_{3,4}$ of 3.5/1, indicating that the magnetic field was almost parallel to the substrate surface; the mean angle between the magnetic field and the Mössbauer γ-rays (perpendicular to the substrate) was calculated to be 75°. The intensity ratio $I_{2,5}/I_{3,4}$ decreased with increasing Ar pressure. The angles between the magnetic field and the Mössbauer γ-rays were calculated to be 60°, 49°, 39°, 34°, and 28° for the films produced at 0.1, 0.5, 1, 3, and 10 Torr Ar pressures, respectively. This indicates that the magnetic field tends to become increasingly perpendicular to the substrate surface with increasing Ar pressure. Collisions of Fe atoms with Ar gas molecules reduce the translational energies of the laser-evaporated Fe atoms. When the Fe atoms have high energies, the Fe atoms migrate on the film surface to form α-Fe crystals parallel to the surface. In contrast, Fe atoms with low energies do not migrate on the surface, so that α-Fe crystals grow perpendicular to the surface plane. Consequently, the orientation of the magnetic field of an α-Fe film can be controlled by the Ar pressure.

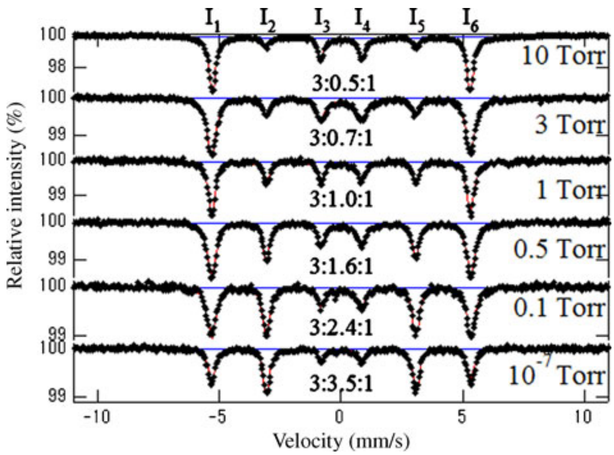

Fig. 1 Mössbauer spectra of α-iron thin films produced by PLD in Ar atmosphere at room temperature. The Ar pressure during PLD and the area intensity ratio $I_{1,6}$:$I_{2,5}$:$I_{3,4}$ are indicated in the figure

The surface morphology of the films was observed by SEM (Fig. 2). The film produced at an Ar pressure of 10^{-7} Torr had a smooth surface (Fig. 2a), whereas the film produced at an Ar pressure of 20 Torr had a rough surface (Fig. 2b). In addition, the cross section of the film produced at 20 Torr was observed through a crack (Fig. 2c). It shows column-shaped crystals aligned perpendicular to the substrate surface, indicating that the crystals mainly grew perpendicular to the surface. This finding is consistent with the Mössbauer spectroscopy results and thus the shape anisotropy of the α-Fe crystal is considered to be the origin of the magnetic orientation.

To estimate the energy of Fe atoms on the substrate, we performed Monte Carlo calculations [6] assuming a low-density Fe vapor. However, the calculation underestimated the Fe atom energy and the calculated energy was almost the same as that above 0.5 Torr, which does not agree with the present experimental results. Therefore, we recalculated using simple thermal equilibrium assuming a high-density Fe vapor; high-temperature Fe vapors and room-temperature Ar gas were mixed and reached the equilibrium temperature in a plume produced by a laser shot. The equilibrium temperature T_{eq} is calculated using

$$T_{eq} = \frac{T_{iniFe} M_{Fe} C_{V_{Fe}} + T_{iniAr} M_{Ar} C_{V_{Ar}}}{M_{Fe} C_{V_{Fe}} + M_{Ar} C_{V_{Ar}}},$$

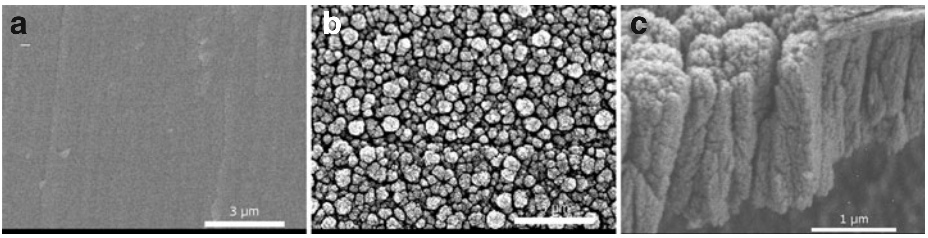

Fig. 2 SEM image of the films produced at Ar pressures of (**a**) 10^{-7} and (**b**) 20 Torr and (**c**) the cross section of a film produced at 20 Torr

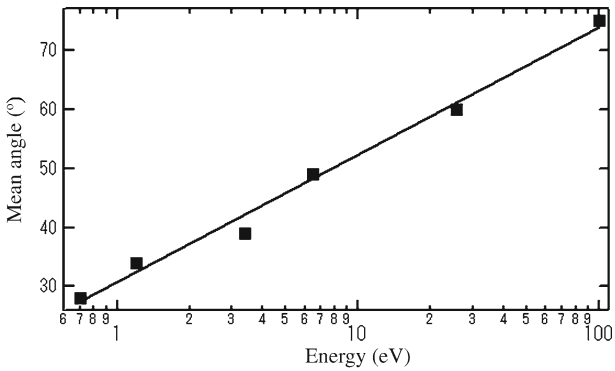

Fig. 3 Single logarithmic plot of the mean angle at each pressure and the energy of Fe atoms calculated by assuming thermal equilibrium

where M_{Ar} and M_{Fe} are respectively the amounts of Fe vapor and Ar gas in the plume, C_{VAr} andC_{VFe} are respectively the specific heats at constant volume of Ar and Fe, and T_{iniAr} andT_{iniFe} are respectively the initial temperatures of the Fe vapor and Ar gas (293 K). Assuming that the Fe atoms have an initial energy of 100 eV, the Fe atoms were estimated to have an energy in the range 0.7 and 100 eV. Figure 3 shows the correlation between the estimated energy of the Fe atoms and the angle of the hyperfine magnetic field. It shows that the magnetic field orientation at each pressure is correlated with the energy.

4 Conclusion

The orientation of hyperfine magnetic field of α-Fe films could be controlled by PLD in an Ar atmosphere. Mössbauer spectra and SEM images indicated that the orientation of magnetic fields was governed by the magnetic shape anisotropy. At high Ar pressures, the Fe atoms had low energies and the hyperfine magnetic fields of the α-Fe films were perpendicular to the substrate surface. At lower Ar pressures, the hyperfine magnetic field of the α-Fe films was parallel to the substrate surface.

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