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Nonlinear strain dependence of magnetic anisotropy in $CoFe_2O_4$ films on MgO(001) substrates

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$CoFe_2O_4$ films were deposited on MgO(001) substrates using pulsed laser deposition with various laser energy densities. We found that the $CoFe_2O_4$ films were grown in the (001) orientation and that the lattice constant of the $CoFe_2O_4$ films was dependent on the laser energy densities. Perpendicular magnetic anisotropy (PMA) was observed, but the lattice strain dependence was not consistent with the phenomenological model in which the PMA is in proportion to the lattice strain. The lattice strain dependence of PMA was compared with an electron theory in which the spin-orbit interaction and the tetragonal crystal field in the electronic state of a single Co ion are considered. The experimental result does not contradict with the calculation, which shows that the magnetic anisotropy is not proportional to the lattice strain and asymmetric in respect to negative and positive lattice strain. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4853235]

Magnetic materials with perpendicular magnetic anisotropy (PMA) are currently attracting great interest, because these materials can be applied for magnetic recording and spintronics devices. Some research groups have reported that $CoFe_2O_4$ films deposited on MgO(001) substrates have PMA.[1–5] The $CoFe_2O_4$ films can, therefore, be a substitute material for recording layers with PMA in hard disk drives. To optimize the PMA of the $CoFe_2O_4$ films, it is important to clarify the origins of the PMA. One of the possible origins is thought to be strain-induced anisotropy.[5] A phenomenological model of strain-induced magnetic anisotropy suggests that the magnitude of this anisotropy is linearly proportional to the lattice strain.[5,7,8]

In this study, we have fabricated $CoFe_2O_4$ films on MgO(001) substrates using pulsed laser deposition (PLD) method, and have found that the lattice strain can be varied with the laser energy densities. This finding makes it possible to examine the lattice strain dependence of PMA. We compared the experimental result with a recently developed electron theory, where the spin-orbit interaction and the tetragonal crystal field in the electronic state of a single Co ion are considered.

$CoFe_2O_4$ films were deposited on MgO(001) substrates using PLD method at the substrate temperature of 400 °C in background oxygen pressure of 2.0 Pa. A stoichiometric polycrystalline $CoFe_2O_4$ target prepared by a conventional ceramic fabrication technique was ablated by a YAG laser with double frequency ($\lambda = 532$ nm) with the pulse width of 6 ns and the repetition rate of 30 Hz. The energy densities of the laser beam were adjusted between 0.2 J/cm^2 and 1 J/cm^2 by an optical lens. The deposition rates were about 0.3 Å/s and 1.0 Å/s for the films deposited using the laser energy densities of 0.2 J/cm^2 and 1 J/cm^2, respectively. The thicknesses of the prepared $CoFe_2O_4$ films were estimated by low-angle X-ray diffraction (XRD) or cross section scanning electron microscopy to be 50, 280, 300, and 400 nm for the films prepared using the laser energy densities of 0.2 J/cm^2 and 50, 120, 150 nm for the films prepared using the laser energy densities of 1 J/cm^2. Electron probe microanalyzer measurements showed that the composition ratio of cobalt and iron atoms is 1:2. The crystallographic qualities and the lattice constants of the $CoFe_2O_4$ films were investigated using XRD with Cu $K\alpha$ radiation. High-angle-annular-dark-field-scanning-transmission-electron microscopy (HAADF-STEM) and nano beam electron diffraction (NBED) were performed using JEOL JEM-ARM200F. Magnetic hysteresis loops were measured using a superconducting quantum interference device (SQUID) magnetometer.

Typical $\theta - 2\theta$ XRD patterns with the scattering vector perpendicular to the film plane for the $CoFe_2O_4$ films deposited using laser energy densities of 0.2 J/cm^2 and 1 J/cm^2 are shown in Fig. 1. The $CoFe_2O_4$(008) peak was observed for each film. The lattice constants perpendicular to the plane for the $CoFe_2O_4$ films deposited with the laser energy densities of 0.2 J/cm^2 and 1 J/cm^2 were about 8.37 Å and 8.53 Å, respectively, and were not dependent on the thickness. The films had epitaxial relation with the substrates for all the samples. To estimate the in-plane lattice constants of the $CoFe_2O_4$ films, XRD measurements for the $CoFe_2O_4$(111) direction were performed. The in-plane lattice constants of the $CoFe_2O_4$ films deposited using laser energy densities of 0.2 J/cm^2 and 1 J/cm^2 were estimated to be about 8.39 Å and 8.29 Å, respectively.

Figure 2 shows cross sectional HAADF-STEM images of the $CoFe_2O_4$ films deposited using different laser energy densities. These images indicate that the $CoFe_2O_4$ films are grown epitaxially with the substrates and that there are no significant defects due to lattice mismatch in both films. The NBED patterns for the $CoFe_2O_4$ films inserted in Fig. 2

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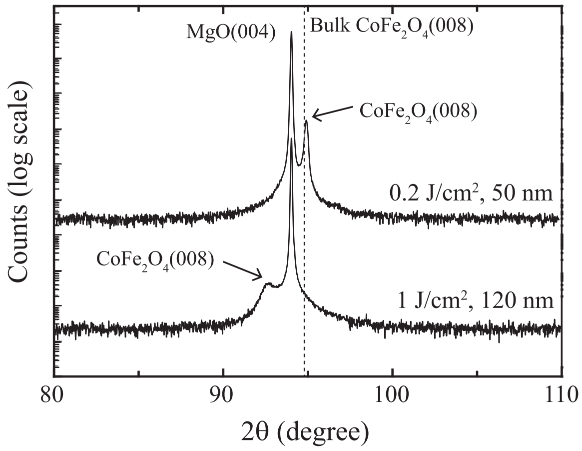

FIG. 1. X-ray diffraction patterns for the CoFe$_2$O$_4$ films grown on MgO(001) substrates prepared with different laser energy densities.

support the lattice strain values of the perpendicular and in-plane directions of the film estimated from the XRD measurements.

Since the lattice constant of bulk CoFe$_2$O$_4$ (8.38 Å) is 0.48% smaller than twice the lattice constant of MgO (4.21 Å), the in-plane lattice constant of the CoFe$_2$O$_4$ films are expected to be larger than that of bulk CoFe$_2$O$_4$. The in-plane lattice constant of the CoFe$_2$O$_4$ films deposited using laser energy densities of 1 J/cm^2 was inconsistent to the expectation. We note that the lattice strain of the CoFe$_2$O$_4$ films deposited using laser energy densities of between 0.5 ∼ 0.8 J/cm^2 was almost the same as that of films deposited using laser energy densities of 0.2 J/cm^2. At the present stage, it is not clear why the difference in lattice strain of CoFe$_2$O$_4$ films occurs as a function of laser energy densities. When the energy densities of the laser beam at the surface of the CoFe$_2$O$_4$ target increase, the velocity of laser-ablated atoms increases and the collision energy of the atoms on the MgO substrates increases. The difference of lattice strain as a function of the energy densities of the laser beam may be caused by the difference in the collision energy of the atoms on the MgO substrates.

Figure 3(a) shows typical magnetic hysteresis loops with magnetic fields applied along the perpendicular and the in-plane directions to the film at 300 K for the CoFe$_2$O$_4$ film grown with the laser energy densities of 0.2 J/cm^2. The saturation magnetization is about a half of bulk CoFe$_2$O$_4$. The similar reduction in the magnetization has been reported by some groups.[3,6] The hysteresis loops clearly show that the CoFe$_2$O$_4$ films grown with the laser energy densities of 0.2 J/cm^2 have PMA. The estimated perpendicular magnetic anisotropy K_u was $+2.2 \times 10^6$ ergs/cc. On the other hand, magnetic hysteresis loops at 300 K for the CoFe$_2$O$_4$ film grown with the laser energy densities of 1 J/cm^2 shown in Fig. 3(b) indicate that the CoFe$_2$O$_4$ film does not have PMA. The estimated K_u of the film was -2.8×10^6 ergs/cc.

Figure 4(a) shows the lattice strain ϵ dependence of magnetic anisotropy K_u of all the prepared CoFe$_2$O$_4$ films with different layer thicknesses. The lattice strain ϵ is defined as $\epsilon = (a_{//} - a_{\perp})/a_0$, where $a_{//}$ and $a_{\perp}$ are the in-plane and the perpendicular lattice constant, respectively, and a_0 is the lattice constant of bulk CoFe$_2$O$_4$. The result in Fig. 4(a) indicates that

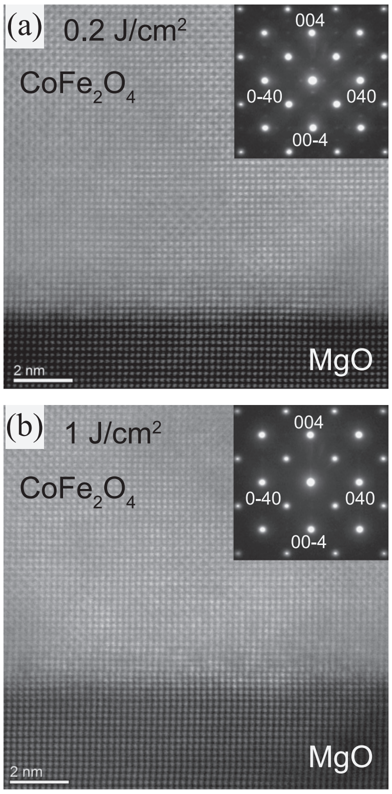

FIG. 2. Cross sectional HAADF-STEM images and NBED patterns for the CoFe$_2$O$_4$ films deposited with the laser energy densities of (a) 0.2 J/cm^2 and (b) 1 J/cm^2.

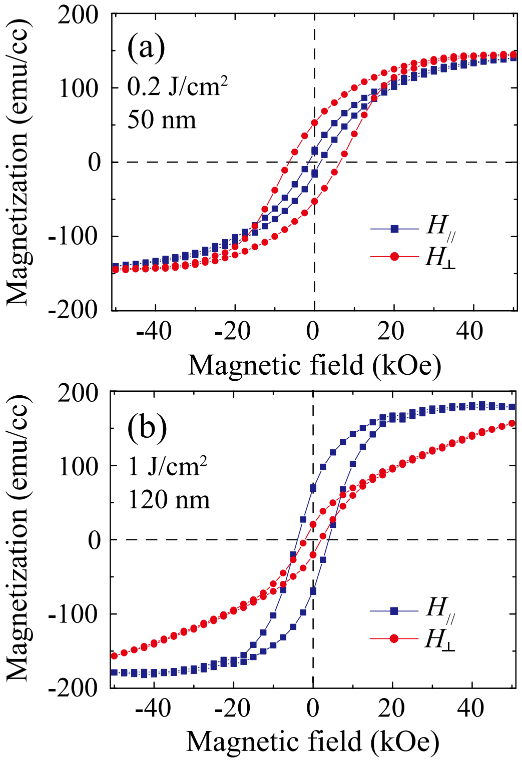

FIG. 3. Hysteresis loops at 300 K for the CoFe$_2$O$_4$ films grown with laser energy densities of (a) 0.2 J/cm^2 and (b) 1 J/cm^2. The loops in red circles and blue squares were obtained for the magnetic field applied along the perpendicular and in-plane directions of the film, respectively.

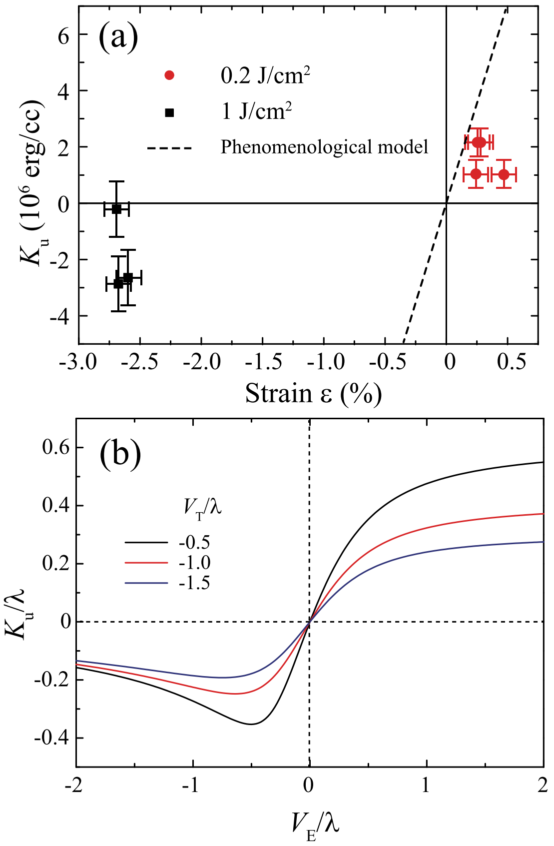

FIG. 4. (a) Lattice strain dependence of perpendicular magnetic anisotropy. The results for the samples with different layer thicknesses are plotted. The dashed line is the calculated result from the phenomenological model. (b) Theoretical calculation estimated by an electron theory where the spin-orbit interaction and the tetragonal crystal field in the electronic state of a single Co ion are considered.

the $CoFe_2O_4$ films which expand perpendicular to the film plane have negative K_u and that the films which shrink perpendicular to the film plane have positive K_u. In the phenomenological model,[5,7,8] $K_u = (1/3)\lambda_{100}(c_{12} - c_{11})\epsilon$. Here, the magnetostriction constant λ_{100} is -590×10^{-6} for $CoFe_2O_4$ and the elastic constants c_{12} is 2.7×10^{11} J/m^3 and c_{11} is 1.1×10^{11} J/m^3.[9] The dashed line which is linear to the lattice strain ϵ in Fig. 4(a) shows the calculated result from this model. The experimental results can not be interpreted using the phenomenological model.

Here, we compare the experimental results with an electron theory for K_u of Co-ferrite films developed recently.[10] The theory takes into account trigonal crystal field (V_T) caused by the three-fold axis symmetry of a single Co ion on the octahedral site, tetragonal crystal field (V_E) due to tetragonal distortion of the film, and spin-orbit interaction (λ). Figure 4(b) shows calculated results of K_u/λ as a function of V_E/λ for several values of V_T/λ. The results show that $|K_u|$ is not proportional to $|V_E|$ but asymmetric with respect to the positive and negative values of V_E. Because the lattice strain ϵ is thought to be proportional to V_E as long as ϵ is small, the phenomenological model may not be supported by this electron theory. Although no experimental data are available for $-2.5\% < \epsilon < 0\%$ at the moment, the experimental results given in Fig. 4(a) may not contradict with the electron theory. Further experimental research is needed to get a deeper insight into the strain-induced magnetic anisotropy.

In summary, the lattice strain dependence of PMA for $CoFe_2O_4$ films prepared by PLD method was investigated. It was found that the laser energy densities of PLD affect the lattice strain of $CoFe_2O_4$ films and that the PMA of $CoFe_2O_4$ films depends on the lattice strain of $CoFe_2O_4$ films. It was also found that the lattice strain dependence of the PMA for $CoFe_2O_4$ films was not consistent with the phenomenological model. The experimental results seem more explainable with the recently developed electron theory, where the spin-orbit interaction and the tetragonal crystal field in the electronic state of a single Co ion are considered.

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