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Elastic constants and magnetic anisotropy of Co/Pt superlattice thin films

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This study is devoted to a correlation between elastic constants and magnetic anisotropy of Co/Pt superlattice thin films. Co/Pt superlattice thin films with various Co–Pt layer wavelengths were deposited on monocrystal silicon substrates by an ultrahigh-vacuum-evaporation method, keeping the volume fractions of the Co and Pt layers unchanged. Their perpendicular magnetic anisotropy ranged between -0.2 and 5.0 MJ/m³. Resonant-ultrasound spectroscopy coupled with laser-Doppler interferometry determined their hexagonal-symmetry elastic constants, which correlate with the magnetic anisotropy; higher perpendicular magnetic anisotropy causes larger in-plane elastic moduli and smaller out-of-plane moduli. The correlation is explained by internal elastic strain associated with lattice misfit at the Co–Pt interfaces. © 2005 American Institute of Physics. [DOI: 10.1063/1.1886900]

Co/Pt superlattice thin films have attracted much attention because of its large perpendicular-magnetic anisotropy (PMA).¹ When Co/Pt superlattice shows PMA, closed-packed planes of Co and Pt layers are epitaxially bonded. Interatomic distance on the closed-packed plane in Pt is larger than that in Co by 10%. Then, Co layers are extended along the in-plane direction, and the in-plane elastic strain is of the order of 10^{-2} .^{2,3} Among possible causes contributing to PMA, previous experimental studies strongly indicate that elastic strain in the Co layer plays very important role. PMA becomes remarkable when the Co-layer thickness is smaller than 4 Å,⁴ which is close to the c -axis lattice parameter in an hcp Co. However, PMA disappears even with the favorable Co-layer thickness when Pt-layer thickness is comparable with or smaller than the Co-layer thickness.⁵ Because the elastic constants of Co layer are smaller than those of Pt layer, elastic strain is more significantly caused in Co layers. This trend becomes remarkable when the volume fraction of the Co layer is small. Kingetsu and his co-authors revealed that the in-plane elastic strain in Co reached 0.05 using the reflection high-energy electron diffraction (RHEED) and indicated that such a large strain governed an important role for PMA.^{6,7}

Extensively large elastic strain affects the elastic constants C_{ij} through lattice anharmonicity. Then, Co/Pt superlattice must show a correlation between the elastic constants and PMA, and determination of the elastic constants can be one of the important approaches for solving the physical mechanism of PMA. However, no report appears studying the relationship between elastic constants and magnetic properties of Co/Pt superlattice despite the large number of publications for this superlattice. Therefore, the purpose of this study is to clarify the correlation between the elastic constants and PMA of Co/Pt superlattice thin films.

Thin films are expected to be elastically anisotropic between the in-plane and out-of-plane directions and often show transverse symmetry with five independent elastic constants denoted by C_{11} , C_{33} , C_{12} , C_{13} , and C_{44} .⁸ This study

determines the C_{ij} of Co/Pt superlattice using resonant-ultrasound spectroscopy (RUS) coupled with laser-Doppler interferometry.^{9,10} (No study applied the RUS method to thin films except for that made by Maynard and his colleagues.¹¹) Mechanical free-vibration resonance frequencies of a film/substrate specimen depend on dimensions, mass densities, and all the elastic constants of the film and substrate.¹² The dimensions are measurable as shown later. The substrate C_{ij} are determined inversely by measuring its resonance frequencies. The film C_{ij} are then determined by measuring the resonance frequencies of the film/substrate specimen and performing the inverse calculation to find the best fits between the measured and calculated resonance frequencies. Details of the measurement setup and correctness of the RUS/laser method are shown in our previous studies.^{13,14}

We prepared Co/Pt superlattice thin films by an ultrahigh-vacuum-evaporation method. Co and Pt layers were deposited on {001} plane of monocrystal silicon substrate, measuring about $6.0 \times 4.5 \times 0.2$ mm³. The pressure before the deposition was of the order of 10^{-9} Torr and it varied during deposition between 0.1 and 9.0×10^{-7} Torr. The deposition rate was 0.3 Å/s for both Co and Pt. At first, 50 -Å-Pt buffer layer was deposited on the silicon substrate. Then, Co and Pt were deposited alternately. The Co–Pt thickness ratio ($d_{\text{Co}}/d_{\text{Pt}}$) was 0.25 . (d_{Co} and d_{Pt} denote the thicknesses of Co and Pt layers, respectively.) We prepared three kinds of superlattice thin films with the bilayer thicknesses $\lambda = 184$, 94 , and 17.7 Å, which were determined by the x-ray-diffraction (XRD) satellite peaks. Figure 1 shows x-ray diffraction spectrum from the film with $\lambda = 94$ Å. It shows

FIG. 1. XRD spectrum of Co/Pt ($\lambda = 94$ Å) superlattice thin film (Cu $K\alpha$).

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FIG. 2. Magnetic hysteresis loops of Co/Pt superlattice thin films with $\lambda = 17.7$ (left) and 184 Å (right). Solid lines indicate magnetic hysteresis loops when the external field is applied along the out-of-plane direction. Dashed lines indicate those for the external field along the in-plane direction.

clearly satellite peaks, which confirms good periodicity. The bilayer thicknesses can be determined by the satellite-peak angles and the x-ray wavelength.¹⁵ Total thicknesses are calculated from the bilayer thickness and the repetition number to be 9250, 9450, and 8900 Å for the $\lambda = 184$, 94, and 17.7 Å films, respectively. We measured magnetic hysteretic loops at 300 K by a superconducting quantum interference device (SQUID) to confirm the epitaxial interfaces and to evaluate PMA as shown in Fig. 2; the Co/Pt superlattice clearly shows PMA when bilayer thickness is 17.7 Å. This result indicates that the Co/Pt superlattice grows epitaxially and includes a larger elastic strain in the Co layer. When bilayer thickness is 184 Å, the easy magnetization direction is parallel to the film surface and PMA disappears.

Figure 3 compares resonance spectrum of the silicon substrate alone and that of the $[\text{Co/Pt}(\lambda=94 \text{ Å})]_{100}/\text{Si}$ specimen. For all specimens, resonance frequencies decreased by 1.0%–1.5% after the deposition. These frequency shifts are larger than the measurement errors of resonance frequencies by two orders of magnitude. Figure 4 shows bilayer thickness λ versus the effective-magnetic-anisotropy energy K_{eff} , which is determined from magnetic hysteretic loops along the in-plane and out-of-plane directions.⁷ K_{eff} indicates the degree of PMA, and positive value means that Co/Pt superlattice shows PAM. K_{eff} decreases as λ increases, which indicates that the magnetization easy axis changes from along the out-of-plane direction to along the in-plane direction with the increase of λ . Figure 4 also shows determined C_{ij} of the Co/Pt superlattice thin films. Error bars (ΔC_{ij}) were determined from the measurement accuracy of the resonance frequencies and that of the film thickness as discussed above. We failed to determine C_{44} because its sensitivity to resonance frequencies was too small. For $\lambda = 184$ Å, we used different specimens, but the differences in the determined C_{ij} were less than 10%. Figure 4 clearly shows correlations between the elastic constants and magnetic anisotropy of Co/Pt superlattice thin films. As λ increases, C_{11} , C_{13} , C_{66} , and E_1 decrease. C_{33} and C_{12} are nearly constant within their error

FIG. 3. Resonance spectra of silicon substrate alone (dashed line) and $[\text{Co/Pt}(\lambda=94 \text{ Å})]_{100}/\text{Si}$ specimen (solid line).

FIG. 4. Dependencies of the effective perpendicular magnetic anisotropy K_{eff} and the elastic constants of Co/Pt superlattice thin films on the bilayer thickness

limits. The ratio C_{33}/C_{11} , indicating elastic anisotropy, increases from 0.97 to 1.4 with λ .

There are two possible reasons for the observed λ dependence of the elastic constants of the Co/Pt superlattice thin films: (i) strain dependence of the elastic constants and (ii) incohesive bonds on columnar grain boundaries.

The XRD measurement indicates that Pt $\langle 111 \rangle$ direction is parallel to the film-thickness direction. Because the closed-packed planes are bonded epitaxially, we assume the closed-packed plane of Co is parallel to $\{111\}$ plane of Pt. The lattice misfit on the two closed-packed planes is as large as 10%. Thus, Pt layers are compressed and Co layers are extended. The in-plane strains in Pt and Co layers are estimated to be -0.018 and 0.086 , respectively, assuming complete bonds at their interfaces. For this evaluation, we used the hexagonal elastic constants of $\{111\}$ -texture Pt, which were obtained from the Hill-average method.¹⁶ For Co, we used monocrystal C_{ij} . An extension of interatomic distance decreases the C_{ij} associated with the extension direction and a contraction increases C_{ij} associated with the contraction direction because of interatomic anharmonicity.^{17,18} The λ dependence of K_{eff} indicates that elastic strains in Co/Pt superlattice are reduced with the increase of λ , because a large elastic strain in Co layer is indispensable to PMA. Therefore, C_{11} and E_1 of Co layer increase and those of Pt layer decrease, as λ increases. The change of the elastic constants due to lattice distortion is expected to be more remarkable in contraction than in extension because the increase rate of the internal energy is larger in contraction. Thus, the change of elastic constants of the multilayer is mainly caused by those of Pt layer and we attribute anisotropy and λ dependence of the elastic constants to the strain-dependent elastic constants.

To reduce the internal strain, dislocations would have to slip along an off-in-plane direction. Seeger *et al.*¹⁹ and Akhtar²⁰ suggested that dislocations could cause $\{11\bar{2}2\}$ $\langle 11\bar{2}3 \rangle$ slip when sufficiently large stress was applied. However, this will be difficult when the Co layer consist of a few atomic layers, because such a case requires dislocation slip across interfaces. Whereas, when the Co layer becomes thicker, the dislocation slips occur and internal strain can be released, which decrease the degree of PMA and the in-plane elastic constants.

Incohesive bonds can also explain the elastic anisotropy. The elastic anisotropy ($C_{33} > C_{11}$) observed in the Co/Pt films show the same trend with that of the CVD-diamond thin films.²¹ The elastic anisotropy of the CVD-diamond films was attributed to incohesive bonds at the boundaries of columnar grains growing along the film thickness. In the case of Co/Pt superlattice, the Co/Pt superlattice may consist of columnar structure and includes incohesive bonds when the thicknesses of Co and Pt are relatively large. Then, observed λ dependence of C_{ij} can be explained by incohesive bonds existing along the columnar structure.

Some studies reported larger C_{ij} in layered films than those predicted by simple rule of mixture, which is called supermodulus effect.²² C_{11} and C_{66} of the Co/Pt superlattice with $\lambda = 17.7$ Å are larger than those of bulks by 4.9% and 6.9%, respectively. However, the significant superlattice effect as reported in the previous studies was not observed.

In summary, we determined anisotropic elastic constants of Co/Pt superlattice thin films by the RUS/laser method, and found bilayer-thickness dependence of the elastic constants (as λ increases, C_{11} , C_{66} , and E_1 decrease) and correlations between elastic constants and PMA. We attribute observed elastic properties to elastic strains caused by lattice misfit and lattice anharmonicity.

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