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Elastic stiffness of $L1_0$ FePt thin film studied by picosecond ultrasonics

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The elastic stiffness of epitaxial and polycrystalline $L1_0$ FePt films is studied by picosecond ultrasonics coupled with x-ray reflectivity analysis, and we find that $C_{33}$ of $L1_0$ FePt is 309 GPa. The morphology of FePt films shows dependence on the film thickness; as the film thickness increases the mazelike structure changes to a continuous film. The elastic stiffness correlates with the morphology change, and it increases as the film thickness increases. When the film thickness exceeds 40 nm, the elastic stiffness becomes independent of the film thickness, and we define the saturated value as $C_{33}$ of $L1_0$ FePt. © 2011 American Institute of Physics. [doi:10.1063/1.3562031]

$L1_0$ FePt shows a chemically ordered face-centered tetragonal structure, which consists of stacks of alternating monatomic layers of Fe and Pt in the [001] direction. It shows high uniaxial magnetic anisotropy in the [001] direction, $K_u = 7.0 \times 10^6$ J/m³ at room temperature,¹ making it a candidate for high-density magnetic recording media. Considerable attention has been focused on $L1_0$ FePt, and the relationship between crystallographic structure, morphology, and magnetic properties have been investigated intensively while varying the fabrication conditions. Despite numerous studies on $L1_0$ FePt, the elastic properties remain unknown. Although the elastic constant is a fundamental parameter to be determined when a material is found or developed, that of $L1_0$ FePt has not yet been measured experimentally. A possible reason for this is the difficulty of fabricating a sufficiently large single crystal of $L1_0$ FePt that the elastic constants can be accurately determined by conventional methods. In the literature, elastic constants of $L1_0$ FePt have been deduced using the modified embedded-atom method (MEAM),² ab initio calculations,³,⁴ and the analytic bond-order potential (ABOP) formalism.⁵ However, the calculated values vary widely depending on the calculation method and the calculation conditions; for example, the component $C_{33}$ of the elastic constant matrix ranges from 242 to 371 GPa.²⁻⁴ Because elastic constants are defined as the second-order derivative of the interatomic potential, this variation implies that a reliable potential has not yet been identified. As a result, other physical properties calculated by these methods could be unreliable. Comparison between calculated and measured elastic constants is a possible way to define a reliable potential, and measuring the elastic constant is indispensable for this task.

In this letter, the elastic constants of epitaxial $L1_0$ FePt film is determined using picosecond ultrasonics coupled with x-ray reflectivity analysis.⁵ This technique is capable of determining the elastic constant of a film thinner than 100 nm using a femtosecond-pulsed laser, and has been applied for several thin films.⁶⁻⁹ In epitaxial growth of $L1_0$ FePt films, a remarkable change in the morphology occurs; with increasing film thickness, isolated particles grow, and coalesce, forming a continuous film.¹⁰ Because macroscopic elastic constants of thin films tend to be different from those of the corresponding bulk materials due to nanoscale defects,¹¹⁻¹⁴ the contribution of these defects to the elastic constants must be considered carefully. In this study, we investigate the relationship between the elastic constants and morphology while varying the total film thickness, and determine the elastic constant of single crystal $L1_0$ FePt. Finally, we compare the experimentally determined elastic constants with reported calculated values.

$L1_0$ FePt films were prepared by depositing a Fe/Pt superlattice on heated substrates referring to the literature.¹⁵,¹⁶ Superlattices were deposited on MgO(001) and borosilicate glass substrates heated to 500 °C. After depositing a 3.6-Å-thick Pt buffer layer, Fe and Pt were deposited alternately. The thickness of each layer was 2.8 Å and 3.6 Å, respectively. The total film thickness was varied between 17 and 73 nm by changing the number of bilayers. The crystal structure was investigated by x-ray diffraction measurements, and the film thickness, $d$, was determined by x-ray reflectivity analyses.¹⁷,¹⁸ The morphology was investigated by the atomic-force microscopy (AFM). For evaluating the effect of the fabrication method on the elastic constants, we also prepared an $L1_0$ FePt film by cosputtering. This film was prepared by depositing Fe and Pt simultaneously via rf-magnetron sputtering onto a MgO(001) substrate at 700 °C. The film thickness was 107 nm. The composition of these films was identified to be Fe₉₂Pt₈₀₋ₓ (x = 50 ± 10) by atomic absorption spectrometry.

The elastic constants of FePt thin films were determined using picosecond ultrasonics. Picosecond ultrasonics is a technique to generate and detect a gigahertz-frequency longitudinal acoustic pulse propagating in the film-thickness direction using ultrashort pulses of light.¹⁹,²⁰ This technique is capable of measuring the round-trip time, $\Delta t$, of the acoustic phonon during repeated reflections between the film surface and the film-substrate interface. The out-of-plane longitudinal elastic constant, $C_{33}$, is determined from $\Delta t$, $d$, and the mass density, $\rho$, by the relation $C_{33} = \rho(2d/\Delta t)^2$. $\rho$ was calculated from the atomic weights and lattice parameters of $L1_0$ FePt. In very thin films, a fundamental standing phonon oscillation was detected instead of multiple reflections of acoustic phonons. In this case, $C_{33}$ is determined from the resonance frequency, $f$, by $C_{33} = \rho(2df)^2$. We used a 15 mW

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800-nm-wavelength pulsed laser for generation and a 5 mW 400-nm-wavelength pulsed laser for detection. Details of our measurement method appear elsewhere.

From the x-ray diffraction spectra in Fig. 1, it was confirmed that polycrystalline $L_1_0$ FePt films were grown on glass substrates. Furthermore, the grains were randomly oriented in the film-thickness direction. On MgO(001) substrates, (001) epitaxial $L_1_0$ FePt films were grown. In-plane x-ray diffraction analyses confirmed the cube-on-cube epitaxial relationship with the substrate; the (100) direction of the $L_1_0$ FePt was parallel to the (100) direction of the MgO substrate (see Ref. 21).

Figure 2 shows a typical reflectivity change as measured by picosecond ultrasonics, plotted as change in reflectivity versus the time delay of the probe light. An intense peak appeared at 10 ps, indicating that an acoustic phonon was generated at this time. Following the initial peak, a train of echo signals was observed ($m=1,2,...,5$), which originates from multiple reflections of the acoustic phonon in the FePt film. $\Delta t$ was determined from the slope of the relationship between the time delay and $m$ (shown in the inset in Fig. 2).

Figure 3(a) shows multiple reflection echoes were observed ($m=1,2,...,5$). Roundtrip time was determined from the slope of the time delay vs $m$ (inset).
mechanism changes from rotation of the magnetization to domain wall displacement. This causes a drastic drop in $H_c$. Similarly, we consider that the structure change from a magelike structure to a continuous film caused the change in magnetic properties at the threshold thickness. Thus, magnetic properties also confirm that for $d > 40$ nm the films are very nearly flawless continuous films.

For $d > 40$ nm, a long-range order parameter $S$ was calculated from the x-ray diffraction spectra to confirm the chemical ordering of the films. $S$ reaches unity for perfectly ordered films, and is zero for a chemically disordered film, which is estimated from the integrated area of the fundamental (002) peak, those of the superstructure (001) and (003) peaks, and other parameters. $S$ of the films was 0.87 ± 0.13, and significant dependence on the film thickness was not observed, indicating that observed fluctuation in $S$ does not affect $C_{11}$ seriously. In Fig. 1, diffraction peaks of the cosputtered film appear at slightly higher angles than those of the multilayered films. The difference corresponds to 0.64% difference in the interplanar distance in FePt(001) direction. Considering the higher-order elasticity, change in the interatomic distance should affect the elastic stiffness. However, the change in $C_{11}$ is estimated to be less than 6%, which is smaller than the measurement error in the resultant stiffness. For these reasons, $C_{11}$ showed dependence neither on the film thickness nor the deposition methods for $d > 40$ nm, and we determine an average value of $309$ GPa, which can be considered the intrinsic $C_{33}$ of $L1_0$ FePt.

Experimentally determined elastic constants are compared to calculated values in Table I. For purpose of comparison, $C_{11}$ was calculated from the reported elastic constants using Hill's approximation. In these reported constants, fluctuation of the values exceeded more than 10%. It should be noted that Müller et al. and Zotov and Ludwig deduced elastic constants using the same method [$ab$ initio calculation using the projector-augmented wave (PAW) with the generalized gradient approximation (GGA)] but their values differ with one another. The discrepancy likely originates from differences in the calculation conditions (such as the choice of $k$-points). These results indicate the difficulty in calculating elastic constants. Among the reported elastic constants, those calculated by $ab$ initio calculation using PAW with GGA and by ABOP are somewhat close to our measured values, which indicates that these methods are suitable for theoretically deducing the mechanical properties of $L1_0$ FePt.

We studied the relationship between the elastic stiffness, morphology, and magnetic properties of $L1_0$ FePt films, and found $C_{33}$ of $L1_0$ FePt to be $309$ GPa using picosecond ultrasonics. The crystallographic orientation of epitaxial films usually depends on that of the substrate on which they are grown. By measuring the out-of-plane elastic stiffness of these films, the other components of the elastic constant matrix could be determined. This study leads to a method for determining the elastic constants of materials of which large bulk samples cannot be fabricated.

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20. See supplementary material at http://dx.doi.org/10.1063/1.3562031 for the in-plane x-ray diffraction spectra and AFM images.

### Table I. Comparison of determined elastic constant (GPa) to calculated values.

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<th>$C_{11}$</th>
<th>$C_{12}$</th>
<th>$C_{13}$</th>
<th>$C_{33}$</th>
<th>$C_{44}$</th>
<th>$C_{66}$</th>
<th>$C_{11}$</th>
<th>$C_{44}$</th>
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<td>Zotov and Ludwig³</td>
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²Reference 2.
³Reference 3.
⁴Reference 4.