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doi: 10.1063/1.3067607

Low damping constant for Co_2FeAl Heusler alloy films and its correlation with density of states

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(Presented 13 November 2008; received 16 September 2008; accepted 3 November 2008; published online 9 February 2009)

Gilbert damping for the epitaxial Co_2FeAl Heusler alloy films was investigated. Gilbert damping constant for the films was evaluated by analyzing the data of ferromagnetic resonance measured at the frequency of 2–20 GHz. Gilbert damping constant for the film without annealing was rather large, while it decreased remarkably with postannealing. Gilbert damping constant for the film annealed at 600 °C was ≃0.001. These behavior of Gilbert damping constant can be well explained by the fact that the density of states calculated from first principles decreases with increasing the degree of *B*2 order. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3067607]

Fast magnetization dynamics of ferromagnets has been studied extensively from the fundamental and technological points of view, and it can be well described phenomenologically by Landau–Lifshitz–Gilbert equation,[1]

$$\frac{d\mathbf{m}}{dt} = -\gamma\mathbf{m} \times \mathbf{H}_{\text{eff}} + \alpha\mathbf{m} \times \frac{d\mathbf{m}}{dt}, \quad (1)$$

where $\mathbf{m}$, $\mathbf{H}_{\text{eff}}$, and γ are the unit vector of magnetization, the effective magnetic field, and the gyromagnetic ratio, respectively. α is Gilbert damping constant, which is the friction coefficient of magnetization motion. The critical current density J_c for the current-induced magnetization switching (CIMS) is proportional to α.[2] CIMS is the key technology for realizing the high density magnetic random access memory, so that the understanding of the mechanism of Gilbert damping is of quite importance. It has been reported recently that α for Co_2MnSi (CMS) Heusler alloy is quite small.[3] In order to clarify the mechanism of Gilbert damping, Gilbert damping for the other Co-based Heusler alloy should be studied. Thus, we investigated Gilbert damping for Co_2FeAl (CFA) films.

50 nm thick CFA films were deposited on single crystalline MgO(001) substrates by means of cosputtering with the elemental targets at RT. Base pressure was better than 2×10^{-7} Torr and Ar pressure was 4 mTorr. The films were capped by 2 nm thick Al layers and the capping layer was naturally oxidized in air. Subsequently, the rapid thermal annealing was performed at temperature (T_a) of 400–600 °C in 10^{-6} Torr. Structural characterization of the films was performed by x-ray diffraction (XRD) with Cu $K\alpha$ radiation and atomic force microscopy. Magnetization measurements were carried out by vibrating sample magnetometer. Other magnetic properties and α were evaluated from the measurements of the broadband ferromagnetic resonance (FMR) with frequency range of 2–20 GHz. All measurements were performed at RT.

Figure 1(a) shows θ-2θ scan of XRD for the films. The reflections of (002) and (004) for *B*2 phase of CFA are only observed. Lattice constant estimated from these peaks was nearly equal to the reported value[4] for any films. Figure 1(b) shows typical example of ϕ scan of the reflection from the (202) planes for the film at T_a=600 °C. The positions of (202) peaks for the CFA film shift 45° from those for (202) peaks for MgO substrate. From these data, the epitaxial relationship for the films was confirmed to be CFA(001)[100]// MgO(001)[110]. T_a dependence of the intensity ratio of (002) to (004) peak is shown in Fig. 1(c). The intensity ratio increases with increasing T_a, and this indicates the degree of *B*2 order and mosaicity are improved. Although annealing temperature is rather high, average surface roughness (R_a)

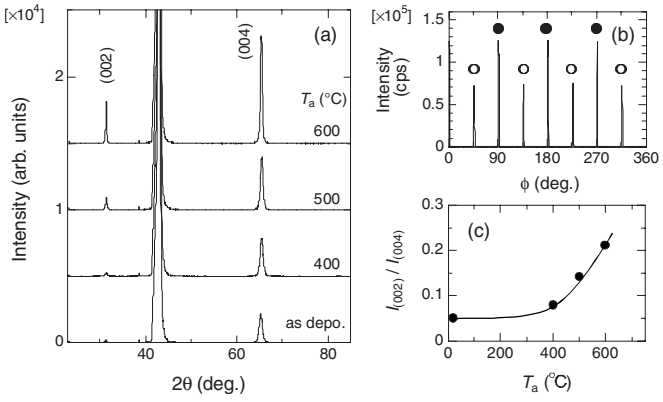

FIG. 1. (a) θ-2θ scan of XRD for Co_2FeAl (CFA) films with various annealing temperature (T_a). (b) The example of ϕ scan of the reflection from the (202) planes for the CFA film (○) and MgO substrate (●) at T_a =600 °C and (c) T_a dependence of the intensity ratio of the (002) to the (004) peak for the CFA films.

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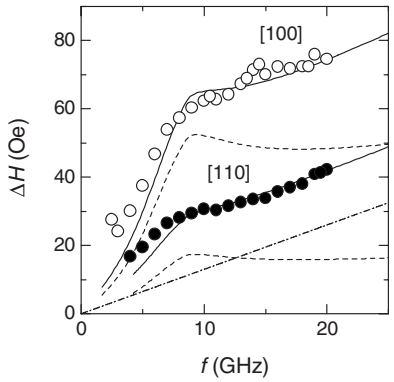

FIG. 2. Typical example of FMR linewidth (ΔH) as a function of frequency (f) for the films annealed at 500 °C with the magnetic field applied in the plane of film along the [110] (●) and the [100] (○) direction. The solid curves are fitted to the experimental data. The dotted and broken lines and the broken curves are the calculated data of ΔH due to the isotropic Gilbert damping and for the anisotropic two magnon scattering, respectively.

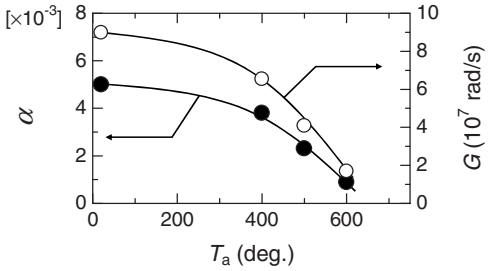

FIG. 3. Annealing temperature (T_a) dependence of Gilbert damping constant (α) (●) and the relaxation frequency (G) (○) for Co_2FeAl films.

was around 0.4 nm for any films. Magnetic moments were almost independent of T_a and were around $\simeq 4.6\ \mu_B$/f.u., which were in accord with the reported value.[4] Magnetization curves were rectangular for any films and the coercivities were around 20 Oe. The magnetocrystalline anisotropy constant (K_1) was estimated from the in-plane angular dependence of FMR. K_1 was $\simeq -8\times10^4$ erg/cc without annealing. It decreased with increasing T_a and became $\simeq -1\times10^4$ erg/cc at T_a=600 °C. The easy direction of the magnetization in the plane of film was ⟨110⟩ for any films.

Figure 2 shows the typical example of frequency (f) dependence of FMR full linewidth at half maximum (FWHM) (ΔH) for the film at T_a=500 °C with applied field along the [100] and [110] directions. In the low frequency regime, ΔH increases rapidly with increasing f and the slope of ΔH versus f along the [100] direction is larger than that along the [110] direction. While, the slope of ΔH versus f changes around 10 GHz, and above 10 GHz the slopes of ΔH versus f for the both direction become almost the same. This nonlinear behavior became more remarkable with increasing T_a. ΔH showed the distinct fourfold anisotropy in the in-plane angular variation in ΔH for the films at f=10 GHz. The minima in ΔH were along the ⟨110⟩ axis, and the maxima in ΔH were along the ⟨100⟩ for any films. This fourfold anisotropy of ΔH also became more remarkable with increasing T_a. Therefore, the nonlinearity for ΔH versus f is closely related to the anisotropy of ΔH. It is well known that ΔH for thin films without any imperfections is proportional to f. Such a nonlinear and the anisotropic ΔH is considered to be due to the two magnon scattering by the line defect or dislocation networks.[5–8] In order to estimate α, we extend the previously proposed formula, which was suggested for isotropic two magnon scattering for thick films,[8] to that for the anisotropic two magnon scattering. Our formula can be expressed as

$$\Delta H = 4\pi\alpha f/\gamma + \Delta H_{\mathrm{mag}}, \tag{2}$$

$$\Delta H_{\mathrm{mag}} = (C_0 + C_1 \cos^2 2\phi)\left|df(H)/dH\right| D_{\mathrm{mag}}(f). \tag{3}$$

Here, C_0 and C_1 are the isotropic and anisotropic two magnon scattering amplitude, respectively. ϕ is the azimuthal angle of magnetization measured from the [100] direction. $D_{\mathrm{mag}}(f)$ is the density of state for the low-energy spin wave, which can be calculated numerically from the dispersion relation for the low-energy spin wave with including the cubic magnetic anisotropy.[8,9] $f(H)$ is Kittel's equation for precession frequency of the uniform mode taking into account the cubic magnetic anisotropy. We performed the fitting to the data of ΔH versus f using Eqs. (2) and (3) with varying C_0, C_1, and α as a fitting parameters. The other parameters were determined from the data of FMR resonance field versus f, and the exchange stiffness (A), which is involved in $D_{\mathrm{mag}}(f)$, was assumed to be 1.5×10^{-6} erg/cm. The calculations are also shown with solid curves in Fig. 2, and these are well fitted to the experimental data using the proper parameters. As seen in Fig. 2, the slope of ΔH versus f above 10 GHz is mostly determined by Gilbert damping, so that the value of α can be uniquely determined.

Figure 3 shows α and the relaxation frequency G for the films as a function of T_a. G was estimated using the relation of $G=\alpha\gamma M_s$, where M_s is the saturation magnetization. γ was determined by analyzing the data of FMR and the values of γ were around 1.9×10^7 rad s^{-1} Oe^{-1} for the films. α and G for the film without annealing are rather large and are comparable to that for Permalloy. While α and G are decreasing remarkably with increasing T_a and are $\sim$0.001 and $\sim 2\times10^7$ rad/s at T_a=600 °C, respectively. These values are comparable to those for FeV films,[10] which are the lowest values of ferromagnetic metals.

Gilbert damping for 3d ferromagnetic metals is generated from the scattering of electrons in d-band perturbed by spin-orbit coupling,[11,12] and the theories predict $G \propto \xi^2 D(E_F)$. Here ξ is the spin-orbit coupling parameter for d-band and $D(E_F)$ is the total density of states (DOS) of d-band at Fermi energy (E_F). The correlation between G and $D(E_F)$ has been never investigated experimentally. For $L2_1$-Co-based Heusler alloys, DOS for the minority spin band is zero or nearly zero and that for the majority spin band strongly depends on the total valence electron number (Z_t). Moreover, $D(E_F)$ is very sensitive to A2 type site disorder. Figure 4 shows the calculated values of $D(E_F)$ as a function of long range order parameter (S) for $B2$ structure. Calculations are done by the *ab initio* simulation code developed by Akai and Dederichs[13] on the basis of the Korringa–

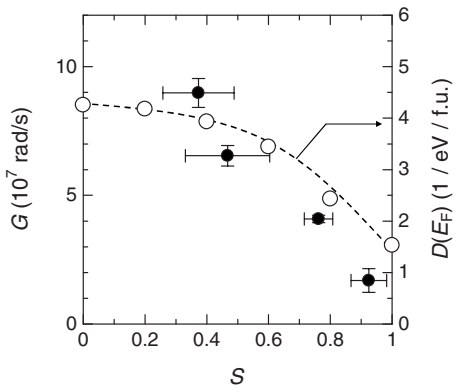

FIG. 4. Relaxation frequency (G) (●) and the calculated total DOS at Fermi level [$D(E_F)$] (○) as a function of long range order parameter (S) for $B2$ order.

Kohn–Rostoker method,[14] where the atomic disorder is implemented within the coherent potential approximation. We adopt the generalized gradient approximation[15] for the exchange and correlation term. The calculated values of $D(E_F)$ decreased remarkably with increasing S, and $D(E_F)$ at $S=0$ (A2) is about three times larger than that at $S=1$ ($B2$). The experimental data of G versus S are also plotted in Fig. 4. S was estimated from the integrated intensity ratio of the peak of (002) on that of (004). The trend of G is very similar to that of $D(E_F)$ within the experimental error, so that T_a dependence of G can be well explained qualitatively by theories based on the band model.

G for $B2$-CFA ($Z_t=29$) in this study is comparable to that for $L2_1$-CMS ($Z_t=29$) (Ref. 3) and is smaller than that for $B2$-Co_2MnAl (CMA) ($Z_t=28$).[16] Therefore, G for Co-based Heusler alloy could be related to Z_t. The theoretical value of $D(E_F)$ for Co-based Heusler alloy also depends on Z_t. The theoretical value of $D(E_F)$ for $B2$-CFA is nearly equal to that for $L2_1$-CMS and is smaller than that for $B2$-CMA in our first principle calculation. This is well understood from the rigid-band picture in half-metallic band structure. On the other hand, G for $L2_1$-Co_2FeSi (CFS) ($Z_t=30$) (Ref. 17) was higher than that for $B2$-CFA in this study although the value of $D(E_F)$ for $L2_1$-CFS calculated using the LSDA+U method[18] is smaller than that for $B2$-CFA in Fig. 4. It should be noted that the value of $D(E_F)$ for $L2_1$-CFS is rather large if U is not taken into account.[18] Further studies are needed for clarifying the correlation between Gilbert damping and DOS for Heusler alloys.

We fabricated (001)-oriented $B2$-CFA epitaxial films and Gilbert damping for the films was investigated. The values of α and G for the film without annealing were large, while those for the film annealed at 600 °C were significantly small ($\alpha \sim 0.001$, $G \sim 2\times 10^7$ rad/s). T_a dependence of G can be well explained by the fact that $D(E_F)$ calculated from first principles decreases with increasing the degree of $B2$ order.

This research was partly supported by Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science, and Technology (MEXT) of Japan and the NEDO grant.

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