Study and tailoring spin dynamic properties of CoFeB during rapid thermal annealing

Yaping Zhang,1,2 Xin Fan,1,a) Weigang Wang,1,3 Xiaoming Kou,1 Rong Cao,1 Xing Chen,1 Chaoying Ni,3 Lijing Pan,2 and John Q. Xiao1,b)
1Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, USA
2Department of Physics, University of Science and Technology Beijing, Beijing 100083, People’s Republic of China
3Physics and Astronomy, Johns Hopkins University, Baltimore, Maryland 21218, USA

Received 15 November 2010; accepted 6 January 2011; published online 26 January 2011

We studied the real-time evolution of magnetic dynamic and static properties of 20 nm CoFeB thin film during annealing at 380 °C. The ferromagnetic resonance linewidth quickly reduces by 30% within 300 s annealing, and monotonically increases upon longer annealing. The magnetic static coercivity shows similar trend. The underlying physical relation between linewidth and anisotropy can be connected by the two-magnon scattering theory. By doping of Nb into CoFeB films, the damping was maintained at a low value within 2000 s annealing. This method to tailor the dynamic properties of CoFeB may benefit the development of magnetics and spintronics based microwave devices. © 2011 American Institute of Physics. [doi:10.1063/1.3549188]

CoFeB alloy is the most common electrode used in magnetic tunnel junctions (MTJs) with crystalline MgO barrier.1–7 As deposited CoFeB layer is amorphous, which serves as a buffer layer to grow crystalline (001) MgO barrier. Upon post thermal treatment, typically done at 350–500 °C from seconds to hours,8–10 CoFeB electrodes crystallized along (001), leading to half-metal-like electronic density of state cross CoFeB/MgO/CoFeB tunnel junction. Consequently, large tunnel magnetoresistance (TMR) are observed. Recently, a rapid thermal annealing process has been demonstrated to increase the CoFeB-based TMR to 200% within 10 min thermal treatment.8 More recently, there are also increasing interest to understand the magnetic dynamics in CoFeB due to the rapid development in spin torques transfer phenomena in MTJs.11,12 One of the common methods to characterize the microwave properties is to study the ferromagnetic resonance (FMR) spectrum, which carries rich information of magnetic dynamics, such as magnetic anisotropy field and damping. The latter is a very important parameter in most microwave applications. A comprehensive study of the ferromagnetic damping in Fe$_{0.97-x}$Ti$_{0.03}$N$_{x}$ shows that the FMR linewidth is related with anisotropy and microstructure, which varies with nitrogen concentration $x$ ranging from 1.9 to 12.7 at. %.13 Furthermore, it has been shown that the magnetization dynamics in FeCoN systems can be tailored by changing the stress in film14 or by adding other elements such as Si, which affect the sample microstructure.15 However the study related to the annealing time dependence of the magnetization dynamics is lacking.16 In this work, we have performed rapid thermal annealing on CoFeB thin films at 380 °C. The annealing time effect on the magnetic dynamics are studied and the underlying relationship between the annealing time and the damping are revealed. Based on our understanding, we added Nb into CoFeB film to effectively minimize the damping during long time annealing.

All samples were deposited in a magnetron sputtering system. The samples structure is Si/SiO$_2/20$ nm CoFeB/20 nm Ta. After the fabrication, the samples were heat treated at 380 °C in an Ar atmosphere for a different time ranging from 20 to 2000 s. These samples were then characterized using a FMR spectrometer and vibrating sample magnetometer (VSM).

A field-swept FMR spectrum is measured at a fixed frequency of 6 GHz with the dc external magnetic field along one designated direction of the sample. In the as prepared sample, the sample exhibits a strong uniaxial anisotropy induced by stress during film deposition. The designated direction is chosen to be approximately along the uniaxial anisotropy. It is observed that the FMR peak position $H_{\text{res}}$ and the linewidth $\Delta H$ evolve with the annealing time as shown in Fig. 1. The linewidth of the as prepared CoFeB sample is about 38 Oe, which is comparable to the value reported by others.17 During the first 300 s annealing, the linewidth quickly decreases to a minimum, which is 30% less than that of the as prepared sample. The linewidth then gradually in-

---

a)Author to whom correspondence should be addressed: fanxin@udel.edu.

b)Electronic mail: jqx@udel.edu.

FIG. 1. (Color online) (a) FMR peak position vs annealing time. (b) FMR linewidth vs annealing time.
creases with the annealing time. This trend of linewidth evolution with the annealing time is also observed in FeGaB thin films.16 The peak position increases with the annealing time, however, due to the complication in the surface anisotropy and easy axis rotation, is not studied in detail.

There are two stages of structural evolution during the annealing process: stress releasing and crystallization. These processes affect the magnetooisotropy. The stress induced anisotropy has a uniaxial symmetry and the crystallization induced anisotropy has a cubic symmetry. An angle dependent FMR measurement can be applied to distinguish these two types of anisotropies. In the presence of uniaxial anisotropy and cubic anisotropy, the modified Kittel equation has a very complicated form. Since in our experiment, the applied external field is much smaller than the magnetization of the samples, the angle dependent FMR frequency approximately follows

\[ \omega^2 = \gamma^2 M_s [H_{\text{res}} + H_u \cos 2(\theta + \theta_0) + H_k \cos 4(\theta + \theta_0)], \]

where \( \omega \) is the resonance frequency, \( \gamma \) is gyromagnetic ratio, \( M_s \) is the saturation magnetization of the material, \( \theta \) is the angle between the external field and the designated direction, \( \theta_0 \) and \( \phi_0 \) are the angles between the designated direction and the easy axis of uniaxial anisotropy and cubic anisotropy, respectively, as illustrated in the inset of Fig. 2(b). \( H_u \) and \( H_k \) are the effective uniaxial and cubic anisotropy fields.

Three exemplary angle dependent FMR fields for samples annealed for 0, 300, and 2000 s are shown in Fig. 2(a). The curve for as prepared sample only shows a twofold symmetry, indicating the dominant of uniaxial anisotropy. After 300 s annealing, where the linewidth reaches the minimum, the uniaxial anisotropy still dominates but its magnitude is reduced. Further annealing results in an even lower uniaxial anisotropy. However, the cubic anisotropy emerges. The \( H_u \) and \( H_k \) are extracted according to Eq. (1) and shown in Fig. 2(b). It is observed that the uniaxial anisotropy decreases while the cubic anisotropy increases with annealing time. It should be pointed out that the anisotropy extracted here is only an average effect among the randomly oriented polycrystalline grains.

To further understand the relation between the anisotropy and linewidth, it is important to understand the contributions of linewidth. In our system, the linewidth consists of three major parts: inhomogeneous broadening, intrinsic spin orbital relaxation, and extrinsic linewidth described by two-magnon scattering (TMS) theory.18

The inhomogeneous broadening, which is frequency independent background linewidth, originates from the inhomogeneities of magnetization and anisotropies at different sample locations. The intrinsic FMR broadening is due to the spin orbital coupling that relaxes the precessional energy into the lattice and dissipates it in the form of thermal energy. The intrinsic FMR broadening can be neglected here because its contribution only dominates in perfect crystal systems. The TMS process describes that the coherent precession is first scattered into different spin wave modes and then dissipated into the lattice. In this condition, the spin waves are quantized as magnons with wave number \( k \), and the uniform precession corresponds to magnon with \( k=0 \). The scattering from \( k=0 \) mode to other spin wave modes is triggered by the grain-grain boundary in the CoFeB films studied here. The linewidth due to grain boundary TMS can be described as

\[ \Delta H_{\text{TMS}} \approx \frac{H^2_0}{M_s} \int \xi^2 C_\xi(\xi) \delta(1 - \omega_k/\omega) d^2k, \]

where \( \omega_k \) is the frequency of the spin wave mode \( k \), \( H_0 \) is the average anisotropy, \( \xi \) is the average grain size, \( \Lambda_{0k} \) denotes the precession ellipticity of the uniform mode and the spin wave modes, \( C_\xi(\xi) = 1/(1 + k^2 \xi^2)^{3/2} \) is the Fourier transform of the grain-to-grain effective field correlation function, the delta function in Eq. (2) means the usable \( k \) modes must be degenerate with the uniform mode frequency. Since \( \int \xi^2 C_\xi(\xi) \delta(1 - \omega_k/\omega) d^2k \) is independent with \( \xi \) and both of \( C_\xi(\xi) \) and \( \Lambda_{0k} \) decreases with \( k \), it can be derived that the integral term in Eq. (2) increases with the grain size \( \xi \). Therefore, in the TMS model, the linewidth increases with both anisotropy fields and grain size. The linewidth evolution with annealing time can then be explained with this TMS theory. The first stage of annealing is stress releasing, resulting in the decrease of the uniaxial anisotropy, hence the linewidth decreases. In the second stage, the cubic anisotropy and grain size increases, giving rise to an increasing linewidth. Due to the small grain size, no systematic study of microstructure is performed. However, it is observed from transmission electron microscopy images that the as prepared CoFeB is amorphous and 1 h annealed sample has a grain size around 7 nm.
Nb compositions are shown in Fig. 4. Compared with un-
sampled. The linewidth versus annealing time with different
cess and linewidth measurement were performed for CoFeB
doped Nb into CoFeB thin films by co-sputtering at the same
application of MTJs where annealing is indispensible.

Besides the dynamic behavior, we also studied the static
magnetization hysteresis with VSM. Figure 3(a) shows three
hysteresis loops of samples annealed for 0, 300, and 4800 s.
In the early stage of annealing, the hysteresis has well de-
dined easy and hard axis. After 4800 s annealing, the hard
axis loop becomes square, and the coercivity of the easy axis
loop and the hard axis loop is very similar as shown in Fig.

Based on the annealing studies, it is learned that the
minimum linewidth can be achieved when the grain size in
the material is small and the stress is low. It is known that Nb
doping in a material can impede the grain growth by accu-
tration varies from 2.8% to 10%. The same annealing pro-
mimulation in the grain boundaries.19 Inspired by this effect, we
doped Nb into CoFeB thin films by co-sputtering at the same
condition as fabricating the CoFeB samples. The Nb concen-
tration varies from 2.8% to 10%. The same annealing pro-
cess and linewidth measurement were performed for CoFeB
samples. The linewidth versus annealing time with different
Nb compositions are shown in Fig. 4. Compared with un-
doped samples, the 2.8% and 5% Nb doped CoFeB films
maintain their linewidths even after 2000 s annealing. For 10% Nb doping, the large linewidth is attributed to the more
inhomogeneity introduced to the system. This study suggests
that Nb can greatly improve the microwave dynamic prop-
erties of the magnetic thin films during long time thermal treat-
ment. This information may be of particular interest to the
application of MTJs where annealing is indispensible.

In summary, an annealing time effect on the static and
dynamic properties of the CoFeB thin film are studied. It is
found that a minimum linewidth and coercivity can be real-
ized at an early stage of annealing. This minimum is attrib-
uted to the quick releasing of stress and the weak TMS
mechanism when the crystalline grain is still small. By dop-
ing Nb into CoFeB, the grain growth is effectively sup-
pressed, which keeps the linewidth as small as 30 Oe after
2000 s annealing. It is expected that the Nb doped CoFeB
can enhance the dynamic properties in microwave applica-
tions, such as spin transfer torque based devices.

This work was supported by the Department of Energy
under Grant No. DE-FG02-07ER46374.

1D. D. Djayaprawira, T. Tsunekawa, M. Nagai, H. Maehara, A. Yamagata,
N. Watanabe, S. Yuasa, Y. Suzuki, and K. Ando, Appl. Phys. Lett. 86, 092502
(2005).
4J. Y. Bae, W. C. Lim, H. J. Kim, T. D. Lee, K. W. Kim, and T. W. Kim, J.
Lett. 90, 212507 (2007).
J. Jordan-sweet, X. M. Kou, Y. P. Zhang, R. Stearrett, E. R. Nowak, R.
8W. G. Wang, C. Ni, A. Rumaiz, Y. Wang, X. Fan, T. Moriyama, R. Cao, Q.
Y. Wen, H. W. Zhang, and J. Q. Xiao, Appl. Phys. Lett. 92, 152501
(2008).
9W. G. Wang, J. Jordan-sweet, G. X. Miao, C. Ni, A. K. Rumaiz, L. R. Shah,
X. Fan, P. Parsons, R. Stearrett, E. R. Nowak, J. S. Moodera, and J.
11G. D. Fuchs, J. C. Sankey, V. S. Priibag, L. Qian, P. M. Braganca, A. G. F.
Garcia, E. M. Ryan, Z. P. Li, O. Ozatay, D. C. Ralph, and R. A. Buhrman,
12C. Wang, Y. T. Cui, J. Z. Sun, J. A. Katine, R. A. Buhrman, and D. C.
14F. Xu, X. Chen, Y. G. Ma, N. N. Phuoc, X. Y. Zhang, and C. K. Ong, J.
15F. Xu, X. Chen, Y. G. Ma, N. N. Phuoc, X. Y. Zhang, and C. K. Ong, J.
17W. J. Gallagher, S. S. P Parkin, Y. Lu, X. P. Buan, A. Marley, K. P. Roche,
Phys. 81, 3741 (1997).
18P. Krivosik, N. Mo, S. Kalarickal, and C. E. Patton, J. Appl. Phys. 101,
083901 (2007).