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Journal Name

RSCPublishing

ARTICLE

Cite this: DOI: 10.1039/x0xx00000x

Received ooth January 2012, Accepted ooth January 2012

DOI: 10.1039/x0xx00000x

www.rsc.org/

Improved Magnetic Damping in CoFeB|MgO with N-doped Ta Underlayer Investigated using Brillouin Light Scattering Technique

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Understanding the role of interfacial roughness on magnetic damping is important for developing advanced spintronics based devices. Using Brillouin light scattering (BLS), here we investigate the interfacial roughness contributing to magnetic inhomogeneity in the technologically important ultrathin CoFeB|MgO heterostructures with Ta and N-doped Ta underlayers. The spin wave frequencies corresponding to uniform precession modes in the BLS spectra for perpendicularly magnetized as well as in-plane magnetized films are investigated. The linewidth shows pronounced dependence on the N-doping in the underlayer. The analysis of linewidth suggests that the magnetic inhomogeneity and the damping reduce significantly by N-doping in the underlayer.

Introduction

With growing need to achieve Perpendicular Magnetic Anisotropy (PMA)¹ in ultrathin films for spintronics based devices, it is important to develop solid understanding of magnetic inhomogeneity²⁻³ and damping⁴⁻⁵ in them. A high degree of magnetic uniformity over large length scale in ultrathin film is beneficial for device applications where interface magnetic properties play key role. Perpendicularly magnetized Ta|CoFeB|MgO heterostructure⁶ is attracting much attention recently as it constitutes the heart of magnetic tunnel junction and the recently discovered three terminal devices.⁷ It is known that in Ta|CoFeB|MgO heterostructure the PMA arises owing to electronic effect at CoFeB|MgO interface.⁶ The choice of underlayer is also known to control the strength of magnetic anisotropy in these heterostructure.⁸ It has been found recently that the PMA of Ta|CoFeB|MgO film can be tuned by changing the N-flow rate during Ta underlayer deposition.⁹ Specifically, CoFeB|MgO films grown on optimally N-doped Ta underlayer has larger interface anisotropy than the film deposited on Ta underlayer. Due to complicated sample preparation for the direct structural characterization when the CoFeB thickness is ~ 1nm (required in achieving PMA)¹⁰ it is quite difficult to investigate the structural inhomogeneity which in turn leads to magnetic inhomogeneity.^{3, 11-15} One of the main contributors of magnetic damping in the thin film is magnetic inhomogeneity whose precise control and understanding is worth to address. A reliable and non-destructive method to investigate magnetic inhomogeneity is the investigation of spin wave. Brillouin Light Scattering (BLS) is a powerful technique to study spin wave frequencies in magnetic thin films and multilayers.¹⁶⁻²⁰ Earlier in Giant Magnetoresistive (GMR) structures spin wave spectra studies using BLS allowed deep understanding of interlayer exchange coupling²¹. In a typical BLS measurements the laser is focused onto a sample, and the photons are inelastically back-scattered by the spin waves¹⁸, Due to momentum and energy conservation the events comprising of annihilation of magnons propagating towards the incoming laser beam (the anti-Stokes process), and creation of magnons propagating in the opposite direction (the Stokes process) take place. The linewidth of the BLS spectra contains the information of magnetic inhomogeneity^{11, 14-15} and in turn the magnon lifetime.²⁻³ Earlier, the linewidth of the spin wave spectra in perpendicularly magnetized ultrathin films has been used to explain magnetic inhomogeneity both qualitatively and quantitatively.11, 15, 22

Using BLS technique here we investigate thermal spin waves in Ta(N)|CoFeB|MgO films. Peak frequency corresponding to uniform precession mode for zero wave vector is detected. The peak frequency varies with external applied field consistent with earlier reported results of perpendicularly magnetized and in-plane magnetized thin films. The magnetic inhomogeneity in these films is investigated by analyzing the linewidth of the BLS spectra. Furthermore, it is shown that the

use N-doped Ta underlayer in CoFeB|MgO heterostructure results in improved magnetic uniformity.

Experimental Details

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Thin films were grown by dc/rf magnetron sputtering in ultra high vacuum (base pressure ~ 2×10^{-7} Pa) onto thermally oxidized Si [100] substrates (SiO₂ thickness is 100 nm). Ta and Co₂₀Fe₆₀B₂₀ layers were grown using dc power while MgO was deposited using rf power in Ar atmosphere. Nitrogen doping in the Ta underlayer was performed by mixing N₂ with Ar during Ta deposition. Based on the flow rate of N₂ and Ar during sputter deposition of Ta, we define a quantity $Q = S_{N2}/S_{Ar} + S_{N2}$. Q was varied up to 9% with Q = 0 corresponding to pure Ta underlayer. The film stacks investigated in the present study are Sub|1 Ta|1 CoFeB|2 MgO|1 Ta, Sub|4 TaN(Q~1%)|1 CoFeB|2 MgO|1 Ta and Sub|4 TaN(Q~9%)|1 CoFeB|2 MgO|1 Ta (digits represent thickness in nm). The films were subsequently annealed at 300 °C for one hour with no magnetic field applied during the process. The static magnetic properties of these films were measured using Vibrating Sample Magnetometer (VSM). The detailed film characteristics (magnetic dead layer thickness and interface anisotropy) have been described elsewhere.⁹ As the aim of this work is to understand the magnetic inhomogeneity of ferromagnetic layer originating mainly due to underlayer/CoFeB interface, so we fix the CoFeB layer thickness to 1 nm. The magnetic dead layer thickness, if any, present in the stack will not be important for investigating magnetic inhomogeneity. Backscattered geometry was selected for performing BLS study of thermal spin waves. The main constituent of the set up is a Sandercock-type six-pass tandem Fabry-Perot interferometer.²³ A laser beam of wavelength 532 nm (from solid state laser) is focused on to the sample and the inelastically scattered light from the sample due to interaction between incident photons and magnons is analyzed to determine spin wave frequency. Here the frequency shift of the scattered beam from the incident laser frequency corresponds to thermally excited magnon frequency. An in-plane magnetic field H up to 3.1 kOe (maximum in-plane magnetic field that can be applied in our BLS setup, which remains stable over the measurement period of each BLS spectrum, which is typically several hours) is applied parallel to the sample surface and perpendicular to the plane of incidence of light. For all these samples the BLS spectra is measured for k = 0 wave vector. Further details of BLS set up may be found in Ref.²⁴

Results and Discussions

Figures 1(a)-(c) show the hysteresis loops measured using VSM for in-plane and out-of-plane directions for three different Q values. The nominal CoFeB film thickness was fixed to 1 nm for all the measurement shown here. The saturation magnetization was calculated by dividing the measured moment value by volume of the ferromagnetic film considering its nominal thickness. Effective anisotropy for the film was estimated by taking the areal difference between the in-plane and out-of-plane hysteresis loops. The positive K_{eff} corresponds

to out-of-plane easy axis for M whereas the negative K_{eff} corresponds to in-plane easy axis. In Fig. 1(d) we plot the saturation magnetization (left axis) and K_{eff} (right axis) as a function of Q. With increase in Q the M_s increases monotonically. The K_{eff} increases for $Q \sim 1\%$, however it reduces to negative value corresponding to in-plane easy axis for $Q \sim 9\%$. In Fig 1(e) we show the relative change in M_s when the temperature is lowered to 100K for Q~1% and Q~ 9% films. We observe 42% and 38% increase in M_s for Q~1% and $Q \sim 9\%$ films at 100K. Earlier for Q ~0% 32% increase in M_s has been reported²⁵. From these studies we understand that these films are thermally robust within the temperature cycling between 300K to 100K.

In Fig. 2(a) we show the measurement schematic for BLS experiment. The laser light impinges at 90° on the sample (parallel to sample normal) thus enabling the measurement for k= 0 wave vector. An in-plane magnetic field H up to 3.1 kOe was applied parallel to the sample surface and orthogonal to incident laser beam. In Fig. 2(b) we show the Lorentzian fit to a typical elastic peak of the BLS spectra to get an estimate of instrumental linewidth broadening²⁶. From the fit we obtain the value of linewidth broadening ~ 0.4 GHz which gives the measure of lower limit of the linewidth in our BLS spectra. The typical BLS spectra obtained for the film stack are shown in Figs. 2(c)-(e). Note that we see the peaks corresponding to the uniform precession mode in all cases due to magnon creation (Stokes) and magnon annihilation (anti-Stokes) resulting from spin wave excitation.



Figure 1: (a)-(c) In-plane and out-of-plane magnetic hysteresis loops plotted in same panel for $Q \sim 0$, 1 and 9%. Inset in each panel shows the schematic diagram of the stack. Digits in the schematic correspond to thickness in nm. (d) Saturation magnetization (left axis, square) and effective anisotropy (right axis, circle) plotted as a function of N-doping in the underlayer. (e) Relative change in the saturation magnetization plotted as a function of temperature for $Q \sim 1\%$ and $Q \sim 9\%$.

Negligibly small difference is found in the intensity, peak frequency and linewidth between Stokes and anti-Stokes peaks at any applied field. The magnitude of the observed frequency

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is related to the effective magnetization (M_{eff}) .²⁷ For out-ofplane magnetized films the $4\pi M_{eff}$ is estimated using Eq (1)²⁷:

$$4\pi M_{eff} = 4\pi M_s - \frac{2K_{eff}}{M_s} \tag{1}$$

We use the M_s and K_{eff} values extracted from the VSM measurement. For Ta and TaN (1%) underlayer films we obtain $4\pi M_{eff}$ of -5.5 kOe and -6.5 kOe, respectively from Eq. (1). The larger peak frequency observed in the case of TaN(1%) is due to larger effective anisotropy and M_{eff} of this film. The BLS peaks as shown in Figs. 2(c) - (e) are fitted using Lorentzian function and the peak frequency and the linewidth (full width at half maximum, FWHM) are obtained. In Figs. 3 (a) and (b) the peak frequency and the linewidth obtained from the anti-Stokes



Figure 2: (a) Schematic for BLS measurement geometry. (b) Lorentzian fit to a typical elastic peak in the BLS spectra. (c)-(e) BLS spectra of the Ta(N)|CoFeB|MgO films at different applied fields for $Q \sim 0$, 1% and 9% underlayer. Field values are mentioned for corresponding spectra.

peaks as a function of the applied in-plane magnetic field are shown for all three films.

From Fig. 3(a) we notice that the films with Ta and TaN(1%) underlayers show a decrease in frequency with the increase in applied magnetic field. For the PMA film, it is known that on increasing the strength of applied in-plane magnetic field the peak frequency first decreases and reaches a minimum at critical field followed by a subsequent increase. The angle between the magnetization and the film plane is determined by the competition between the perpendicular anisotropy and the applied in-plane field.²⁷ The critical field corresponds to the field at which the magnetization is oriented along the film plane at particular applied field. In contrary, the film with TaN(9%) underlayer shows an increase in the frequency with the increase in applied field, a signature of inplane magnetized ferromagnetic thin film.

To further understand the dependence of the peak frequency on applied in-plane magnetic field in perpendicularly ARTICLE

magnetized films (Ta and TaN(Q~1%)) the data is fitted using Eq. 2. 28

$$\left(\frac{\omega}{\gamma}\right)^{2} = (H_{C}^{2} - H^{2}) + \frac{4\pi M_{S} N_{\prime\prime\prime}}{H_{C}} (H_{C}^{2} + H^{2})$$
(2),

where $N_{//}$ is the demagnetization factor, $\gamma = \gamma_e g/2$ is the gyro magnetic ratio, $\gamma_e = 1.759 \times 10^7$ Hz/Oe is the value for free electron, g is the spectroscopic splitting factor and H_c is the critical field²⁸. Considering the film to be isotropic within the film plane we can assume $N_{//} \equiv N_{xx} \approx N_{yy}$. As the demagnetization factors satisfy $N_{xx} + N_{yy} + N_{zz} = 1$ and the films have thickness ~1 nm, we may assume $N_{zz} = 1$ and $N_{//} \approx 0$. The fits using Eq. 2 for Ta and TaN (1%) underlayer cases are shown using red solid curves in Figs. 4(a) and (b). The intercept on x-axis made by the red curve represents the critical field at which the peak frequency takes minima. The critical field obtained using the fit nearly matches to the saturation field for the hard axis loop estimated using VSM. From the fit we obtain $H_c \approx 4.8$ kOe for Ta and $H_c \approx 6.6$ kOe for TaN(1%) underlayer films.



Figure 3: Variation of peak frequency (a) and linewidth (b) with applied magnetic field for all three film stacks.

In Fig.4(c) we show the dependence of frequency on applied field for the in-plane magnetized TaN(9%) underlayer film. The increase in frequency with the increase in bias field is fitted using standard Kittel Eq. $(3)^{28}$ as shown by the red solid curve in Fig. 4(c):

$$f = \frac{\gamma}{2\pi} \left(\left(H + \frac{2K}{M_s} \right) \left(H + \frac{2K}{M_s} + 4\pi M_{eff} \right) \right)^{1/2}$$
(3).

Here *K* is the in-plane anisotropy. Here, we use g = 1.9 and K = 0 and determine $4\pi M_{eff}$ as fitting parameter. The obtained value of $4\pi M_{eff}$ from the fit is 2.2 kOe. From above fits in Figs. 4(a)-(c) we infer that peak frequency dependence on applied magnetic field for Ta, TaN(1%) is consistent with out-of-plane case²⁷ and TaN(9%) is consistent with in-plane case of previously reported literatures.²⁸ For PMA films in our experiment to precisely estimate the minimum frequency in the vicinity of critical field similar to Ref²⁷ is quite challenging. It is primarily due to the fact that the maximum magnetic field achievable in our set-up is well below the critical field. We hope to address this issue in future.

We now discuss below the linewidth of the BLS spectra and its dependence on the Q as well as the applied magnetic-



Figure 4: (a) and (b) Peak frequency dependence on field fitted using standard expression for Ta and TaN (1%) underlayer samples with PMA. The red curve shows the fit using Eq. 2. (c) Plot of frequency dependence on field for TaN(9%) underlayer film. The red curve shows the fit using standard Kittel Eq. 3.

-field (peak frequency). The linewidth shows a drastic reduction with the increase in Q over the whole range of the applied magnetic field (Fig. 3(b)), indicating a significant reduction in the damping parameter with Q. In addition, within the investigated field range (upto 3.1 kOe), the linewidth of the



Figure 5: Variation of the linewidth (FWHM) of the BLS spectra with peak frequency for the samples with (a) Q = 0%, (b) Q = 1% and (c) Q = 9%.

film with Ta underlayer increases by nearly by a factor of two with applied magnetic field whereas the linewidth of films with TaN(1%) and TaN(9%) underlayer show negligibly small increase with applied field (Fig. 3(b)). The behavior of linewidth in a PMA film across the critical field has been earlier investigated for Co/Au film and extremely large linewidth at critical field has been reported.¹⁵ Due to diverging nature of the linewidth when the critical field (4.8 kOe) is approached we observe the significant increase of linewidth for Ta underlayer. For the case of TaN(1%), since the applied inplane field is well below the critical field (6.6 kOe), we do not observe remarkable increase in the linewidth. In order to get

more insight into these results, the linewidth of the BLS spectra for all three films are plotted as a function of the peak frequency in Figs. 5(a) to (c). The sharp decrease in linewidth with increasing frequency for film with Ta underlayer indicates extrinsic contribution (interfacial roughness¹⁴ or structural inhomogeneity¹²⁻¹³) to the magnetic inhomogeneity in this film. In contrary, a nearly constant linewidth for TaN(1%) and TaN(9%) underlayers indicate that the extrinsic factors contributing to magnetic inhomogeneity are suppressed by doping N into the underlayer. It is worth mentioning here that the linewidth data shown in Fig. 3 and 5 are not deconvoluted to remove the contribution for instrumental broadening²⁶. Note that in the linewidth data contribution due to the spectrometer broadening will be uniformly present in the BLS signal for all three films and thus it will not change our interpretation. Despite of presence of instrumental linewidth broadening in the BLS spectra, we are able to extract crucial information of reduction of interfacial roughness in these ultra-thinfilms. An important point to note here, using other characterization techniques e.g., transmission electron microscopy or grazing angle X-ray diffraction to study the interfacial roughness is quite challenging.

It may also be possible that the improved structure related magnetic uniformity in TaN(1%) film assist to some extent in achieving larger PMA in this film as observed earlier⁹. A detailed study of linewidth for large field range for both inplane and out-of-plane direction and for different k vectors will be useful for the detailed quantitative understanding of magnetic inhomogeneity in these films. Nevertheless, the observation of large enough perpendicular magnetic anisotropy $(\sim 6.5 \text{ kOe})$ and reduced spin wave linewidth (damping²⁹) by a factor of 2, that is nearly independent of applied field (spin wave frequency) for the film with $Q\sim1\%$ (N-doped Ta underlayer) is important for application in spin transfer torque magneto-resistive random access memory (STT-MRAM) devices due to the larger thermal stability and smaller write current. In addition, the in-plane magnetized sample (Q = 9%) showed the smallest linewidth (estimated damping of ~0.006 using Ref.²⁹), which would be useful for magnonic devices where a long propagation length of the spin waves are required.

Conclusion

In summary, we have investigated spin wave spectra in Ta(N)|CoFeB|MgO films using BLS technique. Frequency corresponding to uniform precession mode in the BLS spectra of perpendicularly magnetized as well as in-plane film is found. For out-of-plane magnetized sample the spin wave frequency takes minima at a critical field which corresponds to the anisotropy field. The linewidth decreases by nearly a factor of 2, which suggests that the magnetic inhomogeneity reduces with enhanced N-doping in the Ta underlayer. The reduced inhomogeneity in these films by modifying underlayer|CoFeB interface indicates in a large reduction in the value of magnetic

damping, which is important for applications in both spintronic and magnonic devices.

Acknowledgements

We acknowledge the financial assistance from Department of Science and Technology Govt. of India under grant no. SR/NM/NS-09/2011 and S. N. Bose National Centre for Basic Sciences under project no. SNB/AB/12-13/96.

Notes and references

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