

Improvement of the interfacial Dzyaloshinskii-Moriya interaction by introducing a Ta buffer layer

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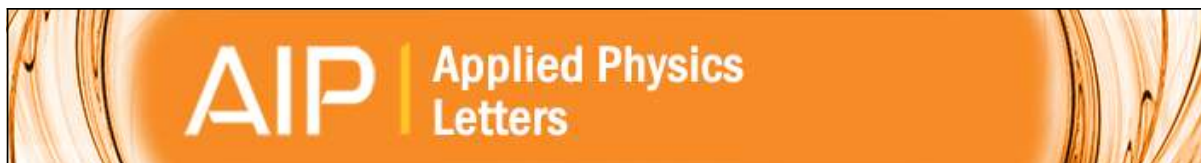
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Improvement of the interfacial Dzyaloshinskii-Moriya interaction by introducing a Ta buffer layer

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We report systematic measurements of the interfacial Dzyaloshinskii-Moriya interaction (iDMI) by employing Brillouin light scattering in Pt/Co/AlO_x and Ta/Pt/Co/AlO_x structures. By introducing a tantalum buffer layer, the saturation magnetization and the interfacial perpendicular magnetic anisotropy are significantly improved due to the better interface between heavy metal and ferromagnetic layer. From the frequency shift between Stokes- and anti-Stokes spin-waves, we successively obtain considerably larger iDM energy densities ($D_{\max} = 1.65 \pm 0.13$ mJ/m² at $t_{\text{Co}} = 1.35$ nm) upon adding the Ta buffer layer, despite the nominally identical interface materials. Moreover, the energy density shows an inverse proportionality with the Co layer thickness, which is the critical clue that the observed iDMI is indeed originating from the interface between the Pt and Co layers.

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In a system with structural inversion asymmetry, spin-orbit coupling at the interfaces introduces an additional asymmetric exchange interaction, which is the so-called interfacial Dzyaloshinskii-Moriya interaction (iDMI).¹⁻⁴ This interfacial nature has recently been experimentally examined because of its massive potential to explore radically different magnetic memory and logic devices based on chiral domain wall dynamics⁵⁻⁸ and skyrmions,⁹⁻¹¹ which are topologically protected vortex- or hedgehog-like spin structures. In order to drive these state-of-the-art technologies on a commercial scale, a larger iDM energy density is strongly required. Recently, it was demonstrated by Brillouin Light Scattering (BLS) that in inversion symmetry broken multilayers (Pt/Co/AlO_x and Pt/CoFeB/AlO_x), the iDM energy density is inversely proportional to the ferromagnetic layer thickness.¹² It indicates that the iDMI is purely originating at the interfaces, as also indicated by recent *ab-initio* calculations.¹³ Moreover, the use of a proper buffer layer can help us to improve the iDMI by introducing an reduced roughness of the interfaces. To increase the interface quality, a buffer layer, especially tantalum (Ta) is widely used in various research fields in magnetism such as perpendicular magnetic recording media composed of Co/Pt, Co/Pd, multilayer system. Since a Ta seed layer can introduce an atomically smooth interface at Pt/Co, a strong interfacial perpendicular magnetic anisotropy (iPMA) can be achieved by the high strain effect,¹⁴⁻¹⁶ and so on.¹⁷⁻¹⁹

In this letter, we experimentally demonstrate that a Ta buffer layer is able to enhance the iPMA and also the iDMI in Pt/Co/AlO_x magnetic multilayer system, which is believed to originate from a better Pt/Co interfaces. In order to investigate these interfacial phenomena, we perform BLS measurements, which is sensitive to the surface spin wave (SW)

excitations. From systematic measurements, we observe the frequency differences between two independent propagating SWs, since these are straightforwardly proportional to the iDM energy density. By introducing a Ta buffer layer, significantly improved iDM energies are clearly observed. Again, the inverse proportionalities of iDMI and iPMA give us strong evidence for the pure interfacial origin of these phenomena.

Our sample consists of Ta (4 nm)/Pt (4 nm)/Co (t_{Co} nm)/AlO_x (2 nm) on a thermally oxidized Si-wafer. All the layers are deposited by using a magnetron sputtering system, and especially, the Co layer is wedged in the range of 1.30–1.80 nm. As depicted in Fig. 1(a), BLS measurements are carried out with strong in-plane magnetic (x -direction in experimentally coordinate system) applied fields up to $H_{\text{ex}} = 0.9$ T. The BLS spectra are observed by using a (3 + 3) multi-pass tandem Fabry-Pérot interferometer, and a p -polarized LASER (300 mW power and 532 nm wavelength) is used as a light source. The back-scattered light is focused from the sample, and the s -polarized lights are passed through the interferometer and collected by the photomultiplier tubes.^{12,20,21} In general, BLS spectra representing Damon-Eshbach surface modes are composed of two propagating SWs signals, the so-called Stokes and anti-Stokes peaks from two different interfaces or surfaces. If the interfaces are identical, two peaks should be located at the same frequency. For non-identical cases, the frequency difference between two propagating SWs exists, which is, e.g., seen in Figure 1(b) showing the BLS spectrum for Ta/Pt/Co/AlO_x with a huge frequency difference ($\Delta f = 2.56$ GHz). In line with earlier results,¹² it reflects that our inversion symmetry broken system contains a significant iDMI.^{12,22-24} More details on the measurements and interpretation are explained in Ref. 12.

As a versatile tool, BLS is able to determine other magnetic properties as well, namely, the anisotropy energy, the

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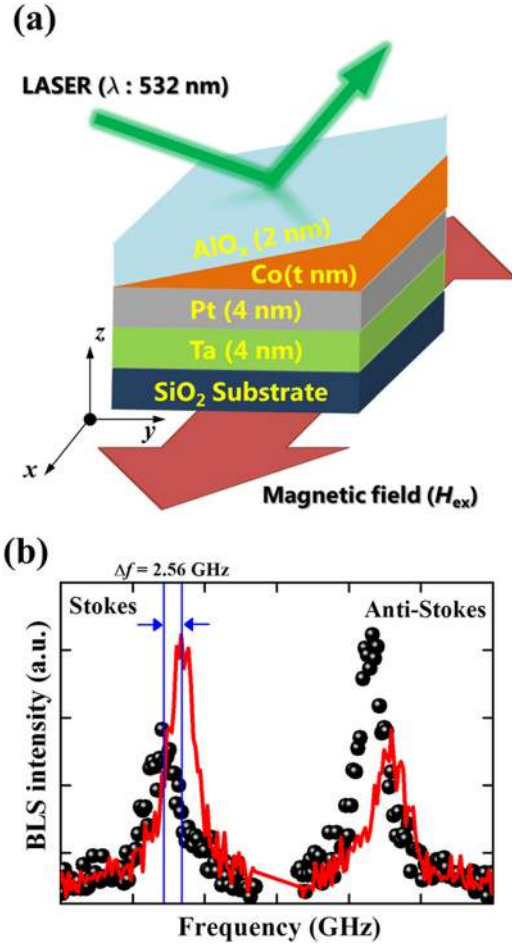


FIG. 1. (a) Schematic for the sample structure and BLS measurement. The sample consists of SiO₂ sub./Ta/Pt/Co/AlO_x, with a wedge shaped Co layer (1.35–1.80 nm). (b) The spin wave spectrum obtained from 1.35-nm thick Co layer with an in-plane magnetic field $H_{\text{ext}} = 0.79$ T. The incident angle is fixed at $\theta = 45^\circ$ (corresponding to $k_y = 0.0167 \text{ nm}^{-1}$). In order to identify the frequency difference (Δf) between Stokes and anti-Stokes, the mirrored curve (red solid lines) is overlapped in the spectrum.

saturation magnetization, and the exchange stiffness constant with the various types of the propagating SWs.^{20,21,25} From BLS measurements, we deduce the effective saturation magnetization value ($M_{\text{eff}} = M_s - \frac{4K_s}{\mu_0 M_s t_{\text{Co}}}$) from the measured

$$\text{SW frequency} \left(f_{\text{SW}} = \frac{\gamma}{2\pi} \sqrt{H_{\text{ex}} \left(H_{\text{ex}} - \mu_0 M_s + \frac{4K_s}{M_s t_{\text{Co}}} \right)} \right),$$

where γ , H_{ex} , M_s , and K_s are the gyromagnetic ratio, the applied magnetic field, the saturation magnetization and the surface anisotropy, respectively.²⁶ In order to extract the change of magnetic anisotropy energy from the measured SWs frequencies in each Co thickness, we used the effective uniaxial anisotropy energy K_{eff} corresponding to the surface anisotropy (K_s) and the volume anisotropy ($-\frac{1}{2}\mu_0 M_s^2$) given by

$$K_{\text{eff}} \times t_{\text{Co}} = 2K_s - \frac{1}{2}\mu_0 M_s^2 \times t_{\text{Co}}. \quad (1)$$

Figure 2 indicates that we observe the linear dependences of $K_{\text{eff}} \times t_{\text{Co}}$ versus t_{Co} with (black squares) and without a Ta buffer (red circles). As shown in Fig. 2, the extrapolated crossing with the y-axis K_s and the slope ($-\frac{1}{2}\mu_0 M_s^2$) are significantly

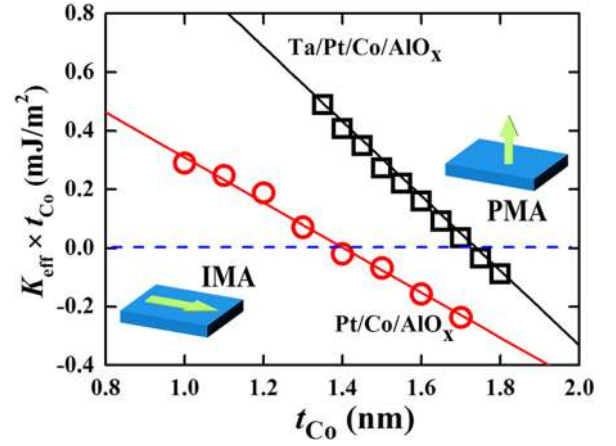


FIG. 2. The $K_{\text{eff}} \times t_{\text{Co}}$ vs. t_{Co} plot with a linear fitting. Above $t_{\text{Co}} > 1.73$ nm (with a Ta buffer layer, black squares) and $t_{\text{Co}} > 1.42$ nm (without a Ta buffer layer, red circles), the effective uniaxial anisotropy becomes negative, which means the easy axis of the sample is in-plane.

enhanced due to the Ta buffer layer. Consequently, the addition of Ta leads to enhance the interface quality of Pt/Co. We will discuss the quantitative changes of K_s (1.10 mJ/m^2) and M_s (1423 kA/m) with Ta buffer layer later on in this paper.

To extract the iDM energy density by using BLS, the first step is observing the frequency differences (Δf) between Stokes and anti-Stokes peaks in BLS spectra. In Figure 3(a), Δf with a buffer (black squares) and without a buffer (red

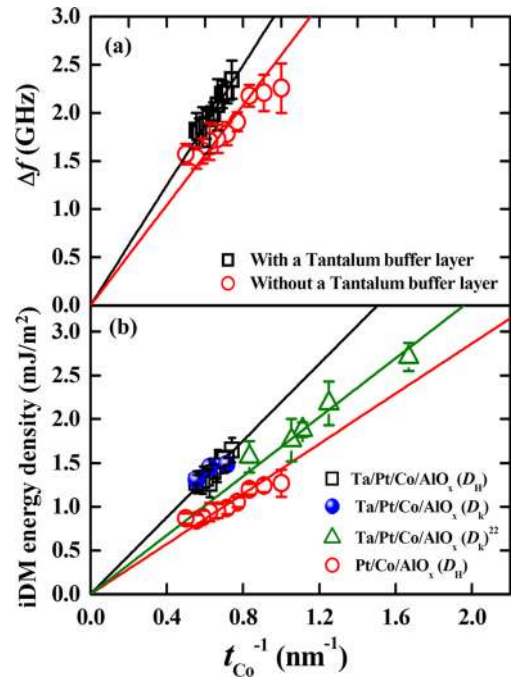


FIG. 3. (a) Δf as a function of t_{Co}^{-1} . Black squares and red squares indicate Ta/Pt/Co/AlO_x and Pt/Co/AlO_x,¹² respectively. For these measurements, the incident angle is fixed at $\theta = 45^\circ$, which corresponds to the $k_y = 0.0167 \text{ nm}^{-1}$. (b) The iDM energy density as a function of t_{Co}^{-1} for the two measurement methods. Black squares and red circles indicate the iDM energy density measured by external magnetic field dependence (D_H) for with (black squares)/without Ta buffer layer (red circles) cases. The blue spheres stand for SW wave-vector dependence results (D_K). D_K are determined from the linear fit of Eq. (2) to the k_y for each thickness ($t_{\text{Co}} = 1.4, 1.6, \text{ and } 1.8 \text{ nm}$). Consequently, black squares, blue spheres and green triangles (Ref. 22) show clearly more improved iDM energy densities, when a Ta is used for buffer layer.

circles)¹² are shown as a function of t_{Co}^{-1} . Here, Δf for each thickness t_{Co} are determined from the field dependent measurements (from 0 to 0.9 T), and the measured Δf should be a constant for all magnetic fields (Ref. 12). Therefore, in Fig. 3, symbols and error bars indicate the averaged values and the corresponding standard deviations of the Δf , respectively. We plot the Δf as a function of t_{Co}^{-1} and it clearly shows the inverse proportionality with t_{Co} . The physical meaning of the inverse proportionality is that a bulk contribution screens the interface effects with increasing t_{Co} . In various magnetic systems, the inverse proportionality to the ferromagnetic layer thickness is a signature of the interface effects such as interface PMA,²⁷ exchange bias,²⁸ the effective field of the interlayer exchange coupling,²⁹ and so on.³⁰ Apart from the observed interfacial nature of Δf , we found that Δf with Ta buffer layers is much larger than Δf without Ta buffer layers, since it is directly linked to the iDM energy density, which is given by

$$\Delta f = \frac{2\gamma D}{\pi M_s} k_y, \quad (2)$$

where k_y and D are the propagating SW k -vector along the y -direction and the iDM energy density, respectively. The SW vector is fixed at $k_y = 0.0167 \text{ nm}^{-1}$ for the field dependent measurements, and it is varied from 0.01 to 0.02 nm^{-1} for the SW wave-vector dependent measurements.

Figure 3(b) shows iDM energy density deduced from Eq. (2) as a function of t_{Co}^{-1} with and without a Ta buffer layer from the magnetic field dependent measurement (D_{H}). We also included the SW wave-vector dependent results (D_{k}) as blue spheres in Fig. 3(b) for selected Co thicknesses (1.4, 1.6, and 1.8 nm), which we obtain from varying the propagating spin-wave k -vector ($0.01 \text{ nm}^{-1} < k_y < 0.02 \text{ nm}^{-1}$). The excellent agreement between two measurement results (D_{H} and D_{k}) implies that our results are independent from possible artifacts as already discussed in Ref. 12.

There are two main issues in this study which we would like to discuss in more detail. First, the iDM energy density with a Ta buffer layer ($D_{\text{H}} = 1.56 \text{ mJ/m}^2$) is noticeably enhanced approximately 58% compared to the absence of Ta ($D_{\text{H}} = 0.98 \text{ mJ/m}^2$) on the same thickness ($t_{\text{Co}} = 1.4 \text{ nm}$). In Ref. 22, the authors have used the same buffer layer and measured iDM energy density by using BLS. The thickness dependent iDM energy densities from Ref. 22 are depicted in Fig. 3(b) (green triangles) and their measured iDM energy densities with a Ta buffer are also reasonably large. Consequently, their results can support our data that a Ta buffer layer is able to improve the iDMI. Second, the iPMA and M_s values are also enhanced by 103% and 29%, respectively, by adding the Ta buffer layer. In order to have a strong interfacial surface anisotropy at the interface between Co and Pt, FCC (111) orientation to induce a high strain effect is necessary. Previous results^{14–16} can clearly support our data that a Ta seed layer can introduce an atomically smooth interface at Pt/Co and then a strong interfacial PMA can be achieved by the high strain effect. Therefore, a Ta buffer decreases the interfacial roughness and then the interface has a strong magnetic anisotropy. As a result, better interface quality will provide stronger spin-orbit coupling,

which is the source not only for iDM interaction, but also for the iPMA and spin polarization of the Pt layer.

In Fig. 3(a), the slopes of Δf values for with and without Ta buffer are similar to each other. However, Fig. 3(b) shows that the slopes of the iDM energy densities are quite different. It is not a surprising result, because M_s is closely related with the exchange stiffness constant A_{ex} , and D should be proportional to A_{ex} .³¹ Therefore, we are able to highlight that the case of a Ta buffer layer gives us a larger M_s ($=1423 \text{ kA/m}$), which is quite close to the bulk value of Co.³² There are two possible scenarios for the large M_s close to bulk Co. First, the improved the interface between Pt and Co layers makes the Co better defined without much intermixing, which should enhance M_s towards the bulk. The second scenario is the proximity effect of the Pt.³³ It is well known that Pt is easily spin polarized and becomes ferromagnet when it is adjacent to the ferromagnetic layer due to the strong spin-orbit coupling and band hybridizations.^{34–36} Therefore, the spin polarized ferromagnetic Pt may contribute to the measured M_s . Without further analysis on systems with a systematic variation of the Pt layer thickness as well (which is beyond the scope of this paper concentrating on iDMI), we are not able to discriminate between the two scenarios.

Finally, we discuss the role of a Ta buffer layer in view of skyrmion formation conditions.^{8,37} The skyrmion phase can be formed when the domain wall energy density, $\sigma = 4\sqrt{A_{\text{ex}}K_{\text{eff}}} - \pi D$, becomes negative, from which we obtain the critical iDMI energy density, $D_{\text{cri}} = 4/\pi \sqrt{A_{\text{ex}}K_{\text{eff}}}$. In our study, we found averagely 58% enhancement of D ; however, K_s also increases by about 103%. In addition, we speculated on an increase of A_{ex} based on the relation with M_s . Even though we enhanced D with a Ta buffer layer, it leads to the enhancement of K_s and A_{ex} , and causes an increase of D_{cri} . Therefore, independent control of D , K_s , and A_{ex} is necessary in order to satisfy the condition for skyrmion formation.

In conclusion, from BLS measurement in Pt/Co/ AlO_x and Ta/Pt/Co/ AlO_x , we obtain that the Ta-buffer significantly enhances the surface magnetic anisotropy K_s , the saturation magnetization M_s , and the interfacial Dzyaloshinskii-Moriya interaction (iDMI). Finally, we emphasize that by engineering the interface quality by introducing a proper Ta buffer layer, we achieved a 58% enhancement of D , despite of the nominally identical interface materials. It implies that there is ample room for improving D by interfacial and structural engineering.

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