Letter

Interfacial potential gradient modulates Dzyaloshinskii-Moriya interaction in Pt/Co/metal multilayers

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The actual mechanisms occurring at interfaces underlying the Dzyaloshinskii-Moriya interaction (DMI) remain a question in nanomagnetism. In this study, we investigate the origin of the interfacial DMI, aiming at estimating how independent the DMI contributions of the two interfaces of a FM layer are and what their relative weight in the effective DMI amplitude is. To this aim, we explore the correlation between the effective interfacial DMI and the metal properties, namely, atomic number, electronegativity, and the work function of the metal M. A clear linear relationship is found between the interfacial DMI and the work function difference at the Co/M interface. This result is strong evidence of the independent DMI contributions of the two interfaces for the chosen Co thickness(1 nm). It also suggests that the Co/Cu interface bears no interfacial DMI. These findings can guide the optimization of the magnetic properties of future devices.

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In recent years, novel chiral magnetic textures such as chiral-Néel domain walls (DWs) [1–3], spin spirals [4], or skyrmions [5] in thin multilayers have been at the core of much research. The interest is twofold: Their fundamental physics and their potential as information carriers in the future generation of spintronic devices, e.g., storage, logic, and neuromorphic devices [5]. One of the key interactions involved in the stabilization of these chiral textures is the antisymmetric exchange known as the Dzyaloshinskii-Moriya interaction (DMI) [6–8] that emerges in systems with broken inversion symmetry and large spin-orbit coupling (SOC). These conditions are realized in thin film systems in which a ferromagnetic layer (FM) is sandwiched between a heavy metal and another metallic (M) or oxide layer [9]. Even if the general conditions for its existence are identified, there are still open questions concerning the microscopic origin, the amplitude, or even the sign of the interfacial DMI. In multilayered systems with FM layers thinner than the exchange length, the two interfaces have to be considered and the measured interfacial DMI corresponds to an effective one (D_{eff}). Therefore, it is difficult to know whether the contributions of the two interfaces simply add or interfere in more complex ways and hence to separate the contribution of each interface. It is, therefore, of utmost importance to solve these questions in order to be able to tune DMI values on demand.

The archetypal experimental systems are Pt/Co-based stacks. This bilayer system provides a large perpendicular magnetic anisotropy [10] and a large interfacial DMI [11]. In most studies, a metal (M) or an oxide (MO_x) layer is grown on top of the FM layer either to generate the next repetition in multilayers or as a protecting capping layer in simple trilayer systems. This layer must guarantee the broken inversion symmetry to avoid the cancellation of the interfacial DMI. An effective DMI amplitude results from the combination of the DMI contributions of the two FM interfaces either reinforcing or competing [12,13]. Our main objective is to understand how the interfaces operate by focusing on the influence of the top Co/M interface on the D_{eff} amplitude in Pt/Co/M systems. This parallels the systematic theoretical work of Jia *et al.* [14], albeit in our case not with monoatomic layer (ML) thicknesses. The values of the effective interfacial energy $D_s = D_{\text{eff}} \cdot t_{\text{Co}}$, ($t_{\text{Co}}$ is the Co thickness) are correlated with properties of the top metal M, namely, the atomic number (Z), the electronegativity (χ), and the work function (Φ) as already proposed by Park *et al.* [15]. These last two parameters are expected to have influence over the interfacial potential gradient (∇V) at the M interfaces [16]. Considering Pt/Co/M with M = Ni, Pd, Ru, Al, Al/Ta, and MoSi , it allows us to explore different d-band filling and a large variety of intrinsic properties and therefore to look for general trends.

The multilayers were grown by dc-magnetron sputtering at room temperature on thermally oxidized silicon substrates covered with a 280-nm-thick SiO_2 layer. The buffer layer is composed by 5 nm Ta, promoting good adherence on SiO_2 and inducing (1 1 1) texture in the 8-nm-thick Pt layer, favoring a strong uniaxial perpendicular anisotropy (typically $K_u \sim 0.8\,\text{MJ/m}^3$ for Co thickness of 1 nm) at the Pt/Co interface. A 3-nm-thick Pt layer is deposited on top of all multilayers in order to prevent oxidation. The saturation magnetization (M_s) and the effective anisotropy field ($\mu_0 H_K$ =

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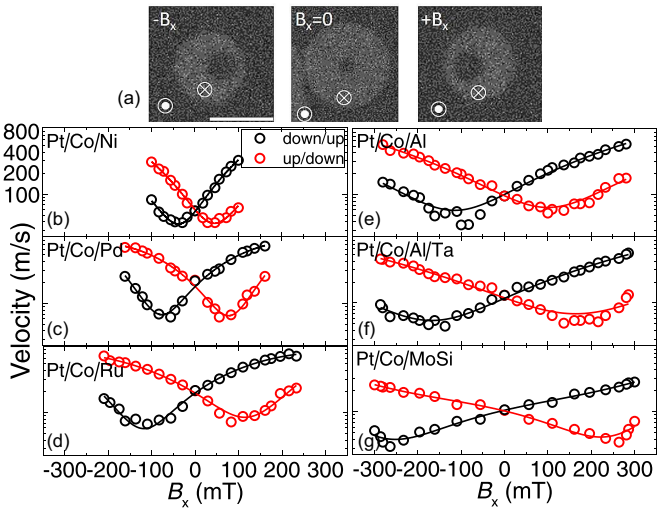

FIG. 1. Determination of the DMI field using asymmetric domain expansion. (a) Expansion of a Néel-type bubble domain in Pt/Co/Al trilayer under out-of-plane magnetic-field pulses (B_z) for different in-plane magnetic fields $B_x = -110$, 0 and 110 mT. Scale bar is 20 μm. (b)–(g) DW velocity vs in-plane field B_x for up/down and down/up DWs propagating in the x direction. The driving B_z = 250 mT field induces a DW propagation in the flow precessional regime. Note that t_{Co} is not the same for the different samples (see Table I).

$2K_{eff}/M_s$) have been measured using standard magnetometry techniques.

We intend to combine results for the amplitude and sign of D_{eff} from asymmetric expansion of bubble domains and Brillouin Light Scattering (BLS). We first present results from the asymmetric expansion of bubble domains measured by polar magneto-optical Kerr microscopy [17–21]. The DWs are driven by out-of-plane magnetic-field pulses (B_z) up to 500 mT and duration down to 30 ns in the presence of a static in-plane magnetic field (B_x). The large out-of-plane fields are chosen to ensure that the DW dynamics are held in the precessional flow regime [22], avoiding the smeared and nonsymmetric DW expansion found in the creep regime [12,17,18]. The differential Kerr images in Fig. 1(a) show the symmetric (asymmetric) expansion of a bubble domain driven by B_z field pulses with $B_x = 0$ ($B_x \neq 0$) for the Pt/Co/Al system. The asymmetric expansion of the bubble domain is a signature of the presence of chiral Néel DWs, whose velocities depend on the relative direction of their internal magnetization and that of the in-plane magnetic field. The velocities of up/down and down/up DWs driven by a fixed out-of-plane field pulse as a function of B_x are shown in Figs. 1(b)–1(g). The in-plane field for which the DW velocity reaches a minimum is the one compensating the field $\mu_0 H_{DMI} = D_{eff}/(\Delta M_s)$ that stabilizes the chiral Néel walls. From this field value, the amplitude of the effective interfacial DMI energy D_s is estimated through

$$D_s = \mu_0 H_{DMI} M_s t_{Co} \Delta, \tag{1}$$

where $\Delta = \sqrt{A/K_{eff}}$ is the DW parameter, A the exchange constant, and K_{eff} is the effective anisotropy energy. Hence a quantitative determination of D_{eff} requires the knowledge of A, which is experimentally challenging for such ultrathin magnetic films [23–25]. We assume $A = 16$ pJ/m according to previous studies [26] on Pt/Co-based samples with similar thicknesses [20,26]. Using this value of A in the D_{eff} estimation from DW motion, values were indeed found to be in best agreement with those measured by BLS. The D_{eff} values obtained in this study for all samples, together with the other magnetic parameters used in the calculations, are listed in Table I. Note that a relatively large error (± 0.2 pJ/m) is considered in order to account for the uncertainty on the exact A value.

Let us now comment on the agreement between these results with a constant A and BLS measurements. For the system with a Ru top layer we find that $D_s = -1.00 \pm 0.20$ pJ/m. This value is slightly smaller than the one determined by BLS performed in Pt/Co(1.7 nm)/Ru in Ref. [27], however, with a thicker Co and hence a larger A ($\approx$ 22 pJ/m). For the Pt/Co/Al and Pt/Co/Al/Ta system, D_s agrees within the error bars with those determined by BLS, however, slightly smaller than the one reported in Ref. [28] in which the samples were epitaxial. Interestingly, D_s differs slightly from the one in Pt/Co/Al, highlighting that the Co/Al interface is electronically and/or structurally impacted by the presence of the top Ta layer. For Pt/Co/MoSi [29], we find $D_s = -0.84 \pm 0.20$ pJ/m, again close to the one from BLS. For the Pt/Co/Pd system, D_s is -0.83 ± 0.20 pJ/m, which is a slightly higher value than those found in Refs. [30,31]. Finally for Pt/Co/Ni, D_s is -0.29 ± 0.2 pJ/m, in agreement with Refs. [32,33]. Note that all these D_s values are negative

TABLE I. Magnetic properties of the different Pt/Co/M trilayers: t_{Co} Co layer thickness, t_M metal thickness, M_s spontaneous magnetization, $\mu_0 H_K$ effective anisotropy field, Δ DW width parameter, $\mu_0 H_{DMI}$ DMI field, D_{eff} effective interface DMI energy density extracted from the DMI field [Eq. (1)] with A=16 pJ/m, D_s effective interfacial DMI energy density, and D_s^{BLS} effective interfacial DMI energy density measured by BLS.

Stacking	t_{Co} (nm)	t_M (nm)	M_s (MAm^{-1})	$\mu_0 H_K$ (T)	Δ (nm)	$\mu_0 H_{DMI}$ (mT)	D_{eff} (mJm^{-2})	D_s (pJm^{-1})	D_s^{BLS} (pJm^{-1})
Pt/Co/Ru	1.1	1.4	1.10 ± 0.05	0.52 ± 0.05	7.5 ± 0.5	110 ± 10	-0.91 ± 0.10	-1.00 ± 0.20	-1.27 ± 0.03
Pt/Co/Al	1.0	1.4	1.23	1.10	4.9	165	-0.98	-0.98	-0.99 ± 0.05
Pt/Co/Al/Ta	1.0	1.4/3.0	1.10	1.30	4.7	200	-1.04	-1.04	-0.87 ± 0.07
Pt/Co/MoSi	0.8	1.4	0.87	1.70	4.7	260	-1.05	-0.84	-0.77 ± 0.10
Pt/Co/Pd	0.9	1.0	1.80	0.38	6.8	75	-0.92	-0.83	–
Pt/Co/Ni	0.7	0.4	0.97	0.45	8.6	50	-0.42	-0.29	–

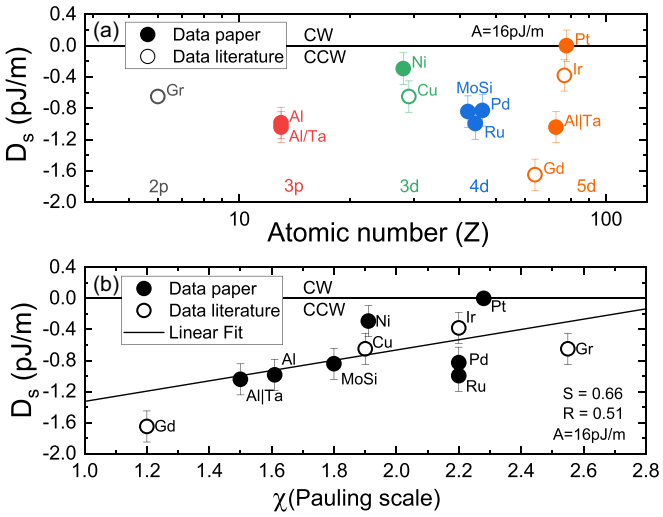

FIG. 2. Dependence of the effective DMI D_s in Pt/Co/M interfaces with (a) the atomic number (Z) and (b) the electronegativity (χ). Bold points correspond to experimental data estimated in this work considering $A = 16$ pJ/m. Open points are values reported from our previous studies

corresponding to a counterclockwise chirality as expected from the one in Pt/Co/M in which D_{eff} is dominated by the large contribution from the Pt/Co interface.

Our main objective remains to investigate experimentally the possible correlation between the estimated D_s values and the properties of the top metals M, namely, the atomic number (Z), the electronegativity (χ), and the work function (Φ). These parameters have been chosen as Z is related to the strength of the SOC, and χ and Φ are related to the expected interfacial potential gradient ∇V, all fundamental ingredients of interfacial effects [9,34,35]. In order to increase the palette of material systems, we have also added to our experimental data some of our previously reported experimental D_s values (obtained using the same MOKE setup), namely, Pt/Co/M, where M = Pt [27], Ir, Cu [28], Gd [20], and Pt/Co/Graphene [21] (open circles in Fig. 2 and Fig. 3).

First we analyze the dependence on the strength of the SOC that is known to increase with the atomic number Z. For our case of interest, the most relevant electrons are the outer ones and in such case, SOC strength proportional to Z^2 is expected [36,37]. In Fig. 2(a), the measured D_s is displayed as a function of Z. Interestingly, we find that the D_s are sorted in different groups depending on their outermost level, i.e., $2p$, $3p$, $3d$, $4d$, and $5d$. No power-law variation with Z can be detected.

As D_s is expected to depend on a charge transfer effect at the interfaces [14], the second parameter of interest is the interfacial potential gradient ∇V. This parameter can be related to the electronegativity χ [38] which is the ability of an atom to attract electrons when it combines with another atom in a chemical bond. The dependence of D_s on χ is scrutinized in Fig. 2(b). A linear correlation is found, however, with a very large dispersion that can be characterized by the Pearson's factor $R = 0.51$. We notice that such a linear behavior has been found in DFT calculations by Jia *et al.* [14] that were, however, performed for MLs Pt(1 ML)/Co(1 ML)/M(1 ML). In this theoretical work, the calculated D_s vary in a range of 5 pJ/m depending on the top M element [14], while experimentally we find a total amplitude variation of only 1.6 pJ/m between Pt and Gd. As in our case, the individual layers are much thicker; it is anticipated that the properties of each interface might change significantly compared to these predictions for atomically thin layers. This discrepancy indeed questions how independent the two interfaces, i.e., Pt/Co and Co/M, can be considered and what their mutual influence is. Recent DFT calculations support the hypothesis that for more than 3 ML of Co (≈ 0.75 nm) the two Co interfaces of Pt/Co/M trilayers may be considered as independent [39]. Accordingly, the behavior of our trilayers is expected to be different from that estimated by Jia *et al.* [14] for 1 ML Co, where the upper Co interface modifies the bottom Pt/Co interface. Within this scenario, we make the hypothesis that the Pt/Co and Co/M interfaces are independent, so that the Fermi level E_F is specific for each element at a given interface.

We finally investigate the dependence of the effective DMI with the work function. To do so, we compare ∇V with Φ which is defined as the minimum energy needed to remove an electron from a surface of a material to the vacuum [40] and is given by $\Phi = -e\phi - E_F$ where $-e$ is the electron charge, ϕ is the electrostatic potential at that surface in vacuum, and E_F is the Fermi level. The Φ values have been collected from the literature [41–43]. We have retained the values of Φ for the (111) surface, as our trilayers are textured in that orientation (when this value was not available, the polycrystalline value has been used). As the Fermi energy is a bulk property, the work function dependence on the surface orientation comes from the electrostatic potential ϕ. Indeed, the electronic charge giving rise to a charge double layer (whose electrostatic effect is ϕ) depends on the orientation of the atomic orbitals and therefore on the surface orientation. The electrostatic potential ϕ varies by a few tenths of eV up to 1 eV in some cases [42]. Importantly in this work, we are considering Co/M interface with no vacuum in between so that the ϕ contribution should ideally be removed. However, as this is not possible, we hence rather consider the work function difference between Co and M: $\Delta\Phi = \Phi_M - \Phi_{\text{Co}}$, expecting $\nabla V \propto \Delta\Phi$. The uncertainty due to the actual value of ϕ is taken into account by error bars of $\Delta\Phi$ coming from the dependence on the surface directions.

In Fig. 3(a), we plot the evolution of the D_s with $\Delta\Phi$ using the values from Table I. The data can be again fitted with a linear relationship, however, with a much better Pearson's parameter ($R = 0.96$) than that found for the evolution with χ, manifesting a much better correlation between D_s and $\Delta\Phi$. Remarkably, the slope of the linear fit (S in the graph) is very similar to that found in Ref. [15]. The observation of a clear correlation between the interfacial DMI and a property ($\Delta\Phi$) depending only on the top Co/M interface supports the assumption that for Co layers with $t_{\text{Co}} \leqslant 3$ ML, the two interfaces Pt/Co and Co/M can be treated as independent. As $\nabla V \propto \Delta\Phi$, we can consider ∇V of the Co/M interface independently added or subtracted to ∇V of the Pt/Co interface. Thus the modulation of D_s depends on the sign of $\Delta\Phi$, leading to an enhancement or a decrease of the effective DMI. Note that the particular case $\Delta\Phi = 0$ corresponds to Pt/Co/Cu [28] for which $\Delta\Phi$ is only 6 meV. From this, we conclude that for Pt/Co/Cu the effective DMI is originating

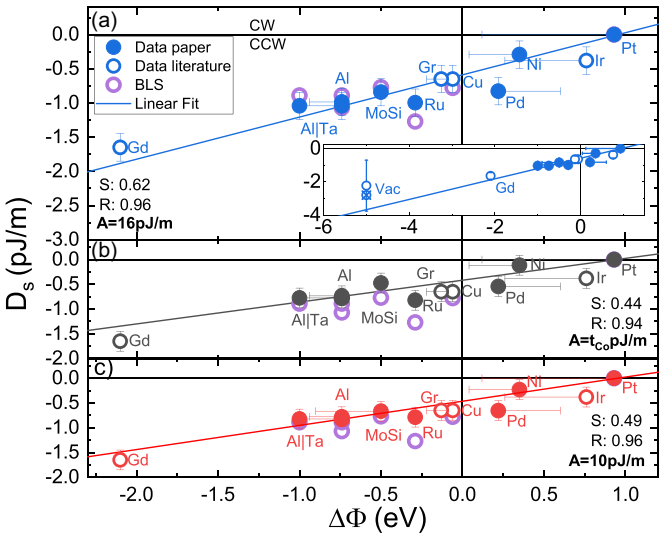

FIG. 3. Dependence of the effective DMI D_s of the Co/M interfaces with work function difference between Co and M ($\Delta\Phi$). The bold points corresponds to experimental data estimated in this work by domain expansion considering in (a) $A = 16$ pJ/m, in (b) a linear dependence of A with Co thickness, and in (c) $A = 10$ pJ/m. The open points are values reported from Refs. [20,21,27,28,44] and the purple open points are values of D_s determined by BLS.

from the Pt/Co bottom interface with almost no additive or subtractive effect from the Co/Cu interface. To further support the independent interfacial contribution reasoning we have added in an inset to Fig. 3(a) the D_s value (upper and lower bounds) extracted from the experimental work of Corredor *et al.* [44] on Pt/Co/vacuum. Note that the upper value of D_s (D_s = -3.7 pJ/m) sits on our linear fit. These values are in good agreement with the calculations for Pt/Co interface ≈ -2.8 pJ/m [represented by $\otimes$ in (a)] [45].

We assumed the fact that single A value might be a strong hypothesis given that the trilayers have different top layer and t_{Co}. In order to demonstrate that our conclusions on the correlation between D_{eff} and different material parameters are not affected by this assumption, two other options have been considered. For the first one [Fig. 3(b)], we consider a linear dependence of A with Co thickness, taking $A = 0$ for 0.5 nm Co (as this is the minimum thickness for which we are able to measure a magnetic signal at room temperature) and $A = 22$ pJ/m for 1.7 nm of Co that we measured by BLS in Ref. [27]. The second hypothesis shown in Fig. 3(c) is to consider $A = 10$ pJ/m. This choice corresponds to the mean value recently estimated experimentally by several techniques in ultrathin Co films very similar to ours [23,24]. Figure 3 shows that whatever the used hypothesis for the A value, the linear dependence of the effective DMI vs $\Delta\Phi$ between the two materials at the Co/M interface is robust. In order to understand the observed trend, we can allude to recent theoretical works connecting the amplitude of the interfacial DMI to the Rashba interaction in a 2D electron gas that is exchange-coupled to a neighboring magnetic layer [46]. However, such Rashba interaction is a combined effect of the asymmetry of the interface, described by ∇V and of the SOC. Thus a theoretical description that goes beyond this simple description would be needed to account for the observed slope of D_s with $\Delta\Phi$.

In conclusion, we have determined the value of D_{eff} in a series of sputtered Pt/Co/M multilayers. The effective interfacial energy D_s is found to clearly depend on the M element in contact with Co in the top interface. Among the different material parameters that we considered, we find a linear dependence of D_{eff} with the work function difference between Co and the top M layer, demonstrating that the interfacial potential gradient at M interfaces plays a crucial role in D_{eff} and that the Pt/Co and the Co/M interfaces can be treated as independent. In the range of thickness considered for the magnetic film ($\geqslant 3$ ML), this experimental observation may provide a means to engineer samples with the desired value of the effective DMI, an interaction that is of major interest in many aspects of today's nanomagnetism.

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