Dzyaloshinskii-Moriya Interaction and its Current-Induced Manipulation

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Abstract

The Dzyaloshinskii-Moriya interaction (DMI) plays a crucial role in the design of advanced spintronic devices. It stabilizes the chirality of domain walls, thereby enabling racetrack type storage devices and allows for the stabilization of topologically non-trivial magnetic textures, such as skyrmions. These textures have various potential applications, including memory and neuromorphic computing. DMI occurs in systems with broken inversion symmetry, which can take on different forms. One form is the bulk DMI, which is usually determined by the intrinsic crystal structure, but a composition gradient may also induce this type of DMI. In thin films, the interfacial DMI is the common way of introducing DMI into a variety of systems. More recently, also a DMI between distinct magnetic layers, known as the interlayer DMI has been introduced. However, the DMI is not only a static material property. In the focus of this review is the potential to manipulate the DMI by electrical fields and currents, which opens up design routes for new devices based on the ability for post-growth control fo the DMI sign and strength of DMI. The effects of currents and fields are laid out both from a theory perspective as well as by reporting the key experiments.

1 Origin of the DMI

Dzyaloshinskii and Moriya first explained the origin of weak ferromagnetism in antiferromagnetic systems, such as α-hematite, due to an intrinsic spin canting mediated by an antisymmetric exchange interaction [1, 2]. Treves and Alexander confirmed this type of interaction in $YFeO_3$ single crystals [3]. Because of the important role played by Dzyaloshinskii and Moriya, the interaction is commonly referred to as the Dzyaloshinskii-Moriya interaction (DMI). The general Hamiltonian for this interaction describes the interaction between two atomic spins $\boldsymbol{S}_1$ and $\boldsymbol{S}_2$: $H_{\mathrm{DMI}} = -\boldsymbol{D}_{12} \cdot (\boldsymbol{S}_1 \times \boldsymbol{S}_2)$. Here, $\boldsymbol{D}_{12}$ represents the DMI vector, which governs the strength and direction of the interaction, as illustrated in Fig. 1. The symmetry constraints on the direction of the DMI vector were given by Moriya [2]. The resulting handedness of positive DMI is inconsistent in the literature, and we refer to the original publication for further details on this topic.

Since the discovery of the DMI, it has been found in various systems with different characteristics. There have been early work on orthoferrites, such as the aforementioned $YFeO_3$ and hematite, where the DMI has been first discovered. In 1980 Fert and Levy proposed that heavy metal (HM) impurities in spin glasses give rise to a DMI due to spin-orbit scattering of conduction electrons [4]. Similarly, HM/ferromagnet interfaces can create an interfacial DMI [5, 6]. This has been demonstrated by the existence of spin helices in Fe/W bilayer systems [7] and later in Mn/W bilayers [8]. This progress has allowed for the implementation of DMI in thin film devices and stabilize Néel-type domain walls in thin films, which enables uniform and highly efficient current-induced domain wall motion [9, 10]. The presence of DMI also leads to the stabilization of magnetic topological particles called skyrmions, which have been first observed in B20 type materials, which display bulk DMI [11] and has opened the field of skyrmionics. We will discuss these different facets of the DMI, starting with bulk the DMI, followed by the interfacial DMI and the interlayer DMI, and ending with a more detailed perspective on the current-induced DMI.

2 DMI in bulk systems (bDMI)

MnSi single crystals exhibit bulk DMI that is intrinsic to their crystal structure, which lacks inversion symmetry [11]. The B20 materials are of particular interest due to their ability to host non-trivial magnetic solitons called skyrmions [11, 12, 13, 14]. These noncentrosymmetric materials have intrinsic DMI and can host skyrmions at low temperatures. However, the intrinsic DMI of these systems comes at the cost of requiring single crystals, as polycrystalline samples would exhibit a non-uniform DMI vector. This is a significant drawback for applications, which the later discussed versions of DMI do not have.

In recent years, a new source of bulk DMI has been discovered in thin film ferrimagnets [15]. These compounds, consisting of rare-earth transition metals, are typically produced through a co-sputtering process, with examples includ-

ing GdCo, GdFeCo, and TbFe. Notably, chiral domain walls were observed in SiN/GdFeCo/SiN nanowires [16, 17], suggesting the possibility of a bulk contribution to DMI. Later, it was confirmed that a DMI exists in these systems, which scales linearly with layer thickness [15]. Scanning transmission electron microscopy and electron energy-loss spectroscopy measurements identified a composition gradient within the GdFeCo layers that was attributed to generate bulk DMI.

Furthermore, Monte Carlo simulations conducted by Liang et al. [18] revealed that a combination of two atom types, A and B, with A acting as a magnetic transition element and B as an element with large spin-orbit-coupling (SOC), generates DMI throughout the full bulk of the material. This finding was experimentally validated by intentionally growing thin films with a composition gradient along the thickness direction. Alloyed films exhibiting a composition gradient along the layer thickness induced a significant DMI that scales linearly with the gradient, with CoPt and FePt alloys being notable examples [19]. In contrast, Fig. 2 displays that alloying Fe or Pt with low SOC materials, such as Cu, did not result in any significant DMI. Interestingly, even the FeGd alloy did not exhibit any sizeable DMI, which contradicts Kim et al.'s observation [15]. The authors attributed this discrepancy to the necessity of SOC from a 5d heavy metal and the inability of 4f rare earths to produce gradient DMI in films with in-plane anisotropy.

Additionally, physical strain applied to a system has been demonstrated to introduce a bulk-like DMI in $La_{0.67}Sr_{0.33}MnO_3$, a centrosymmetric material [20]. The strain gradient along the lateral direction produces an unconventional DMI that has helicoid and cycloid components.

3 Interfacial DMI (iDMI)

The most common source of DMI is interfacial DMI, which is typically found in magnetic thin films. Inversion-symmetry breaking can be generated in the vertical direction by the stacking order of materials. Heavy metals with strong SOC or oxides are combined with magnetic layers in asymmetric stacks [21, 22], such as Ta/CoFeB/MgO [23, 24, 25], Pt/CoFeB/MgO [26, 27, 28], or Pt/Co/Ir [29, 30, 31, 32].

The magnitude of DMI in 3d-5d element combinations has been well described by Belabbes et al. [33] using first-principles calculations. They show that iDMI strongly depends on the hybridization between 3d-5d states around the Fermi level, and the behavior of the DMI is dominated by the Hund's rule filling of 3d shells [33]. Mn displays the largest DMI, with other 3d atoms decreasing in DMI strength the further they stray from the band filling of Mn.

A recent survey of the DMI strength in Pt/Co/metal stacks for a large number of metals reveals a striking linear correlation with the interfacial potential gradient that is originating from the difference in work functions at the Co/metal interface [34]. This already hints at the potential for electrical control discussed in the next paragraph.

The iDMI is especially useful for the stabilization of chiral magnetic textures, such as skyrmions [35, 36, 21]. While B20 materials do host skyrmions, they are mostly stable only at low temperatures. Systems with iDMI can stabilize skyrmions and other chiral structures at and significantly above room temperature [37], which is essential for their use in spintronic applications.

Extensive studies have found iDMI in multiple systems, some of which do not follow the standard heavy metal/transition metal interface. DMI has been found in iron garnets such as TmIG [38, 39, 40] and YIG [41]. Furthermore, chemisorbed oxygen on top of Co/Ni multilayers introduces DMI of similar magnitude and sign as Pd/Ni and Pt/Ni interfaces [42].

Although DMI is typically intrinsically a growth-defined parameter, there are a few ways to tune DMI post-growth. First-principles calculations have shown that tensile and compressive strain can modulate the DMI [43]. Experimentally, strain can be introduced through mechanical deformation [44, 45], piezoelectric substrates [46, 47, 48, 49], and shape-memory-alloy substrates [50]. However, piezoelectric and mechanical strains are well below the 1% strain used in simulations. Nevertheless, strain effects are significant even in experiments, with a 50% increase in DMI observed in Co/Pt multilayers [44] and a suppression of DMI in W/CoFeB/MgO [49]. The relative increase in DMI of Pt/Co/MgO using shape-memory alloys is rather low, despite the strain being about 100 times larger than what is achieved by piezoelectric substrates [50].

Piezoelectric substrates utilize electric fields to manipulate DMI by applying strain to the system. However, the effect of electric fields on magnetic thin films can be directly studied through gating. In Pt/Co/AlOx/HfOx systems, the presence of electrical fields has been shown to result in a change in DMI of $0.14 - 0.26 \pm 0.2 mJ/m^2$ [51]. The change in DMI is largely dependent on the material. Ta/FeCoB/TaOx/AlOx/HfOx also displayed a change in DMI of approximately $0.1 mJ/m^2$ [52], whereas other systems showed significantly smaller values [53, 54, 55, 56, 57].

Recently, ionic liquid gating has been used to induce changes in the magnetic system via the application of electrical fields. In this type of gating, ions can move from the liquid to the capping, and even through the capping [58]. Therefore, in addition to electrical fields, the implantation of ions within the film can induce further changes in the magnetic system. The effect of ionic liquid gating on DMI is significant, but so far, only a reduction of DMI has been observed [58, 59].

4 Interlayer DMI (IL-DMI)

As the interest in the third dimension of magnetism [60, 61] and newly discovered 3D spin textures, such as hopfions [62, 31], continues to grow, the interlayer-DMI (IL-DMI) is gaining increased attention, as it has the potential of stabilizing such structures. This type of DMI can exist in magnetic heterostructures and multilayers and was proposed by Vedmedenko et al. [63]. It has been discovered in synthetic antiferromagnets coupled via an antiferromagnetic Ruderman-Kittel-

Kasuya-Yosida (RKKY) interaction using a non-magnetic spacer [64, 65]. The striking difference of the IL-DMI is, that it favors a perpendicular alignment of spins in two separate magnetic layers. Therefore, it favors a canting of spins in the vertical direction perpendicular to the sample plane in contrast to the canting within the sample plane as for the iDMI. A sketch of this behaviour is shown in Fig. 3. Microscopically the effect also differs from the conventional iDMI as it is mediated by a non-magnetic metallic spacer layer sandwiched between two magnetic layers. It relies on a lateral symmetry breaking compared to the vertical symmetry breaking for iDMI.

In contrast to the other types of DMI, an additional inversion symmetry breaking within the film plane is necessary to introduce IL-DMI. This type of symmetry breaking is growth-induced and can be artificially introduced by oblique columnar growth [65, 66] or static magnetic fields applied during sputtering [64, 67]. A recent study even displayed the presence of IL-DMI in CoPd after hydrogenation [68]. Due to the antisymmetric nature of the effect, the magnitude is largest when one of the layers exhibits perpendicular magnetic anisotropy (PMA) and the other in-plane magnetic anisotropy (IMA). Using this combination of IMA and PMA layers, the strength of the interaction has been characterized using TbFe and Co films separated by a single non-magnetic spacer [69]. The largest IL-DMI was shown by Pt in this system. Furthermore, it was shown that in such a system of perpendicular alignment of magnetic layers, the IL-DMI can be used to realize field-free current-driven switching of both layers, which follows the chiral arrangement governed by the IL-DMI [66].

Even in systems with two PMA layers, the IL-DMI reduces the symmetry, and it has been shown that a current flowing parallel to the DMI vector can further break the symmetry and allow for field-free switching in such systems [67]. Interestingly, the symmetry breaking by the IL-DMI can also stabilize Bloch domain wall chirality in thicker multilayer films, where the increased thickness yields an achiral Bloch component that coexists with a chiral Néel domain wall at the surface, reducing the demagnetization energy. A different population of domain walls has been observed by Lorentz transmission electron microscopy [70].

The IL-DMI is not limited to metallic systems but can also occur when the spacer material is an insulator. It has been shown that NiO sandwiched by Co/Pt layers displays an IL-DMI with a tilting angle that reached 11.5° in the ferromagnetically coupled state [71]. As the interest in IL-DMI continues to grow, its potential for stabilizing three-dimensional structures, as well as its applications for field-free spin-orbit torque switching, become increasingly attractive.

5 Current-induced DMI (CIDMI)

All variants of DMI described so far are resulting from structural and therefore primarily static symmetry breakings. Passing an electrical current or applying a voltage gradient is also breaking the symmetry and can thus modify or give rise to DMI. This offers a very high degree of spatio-temporal control over the

DMI beyond the structural modifications described above.

In a theory view, the spin-orbit interaction can be perturbed by a first-order perturbation to describe the DMI [72]. This results in the ground state spin current being responsible for DMI. It can also be explained as a Doppler shift on the intrinsic spin current caused by spin-orbit coupling under inversion symmetry breaking [73]. Both theories show that a spin current can manipulate DMI. The effect of spin current on DMI can be observed in systems that exhibit perpendicular orientation between spin polarization and magnetization, particularly in magnetic multilayers with perpendicular magnetic anisotropy [72]. However, changes in DMI in Co/Pt systems are small [72], so systems with small intrinsic DMI and/or large current densities are needed.

The use of Ta(5)/CoFeB(0.8)/MgO(2), a system with large perpendicular magnetic anisotropy (PMA) [23] and comparatively small DMI [74, 75, 76], was investigated through current-induced domain wall motion under applied in-plane fields. The experiment was performed on an array of nanowires patterned on a thin film sample [77]. The direction of current-induced domain wall motion is determined by the interplay of spin hall angle and chirality of the Néel domain wall. The spin Hall angle of Ta has been reported to be negative [78, 79, 80, 81], and the current densities used for the measurements are limited to the depinning current on one side and the maximum current that does not induce thermal nucleation events on the other side.

In Fig. 4 (a) we see that, surprisingly, the current-induced motion is opposite to what is expected considering the sign of DMI and current direction. The dependence of DMI on current density is nonlinear and invariant under the current polarity. When data from field-induced domain wall motion, which technically is at zero current, is included, a sign difference in DMI occurs between the current-induced and field-induced method. This has been observed before for Co/Pt/Pt systems [82] and differences between methods to determine DMI have also been observed previously [83, 84]. This discrepancy raises questions that need to be answered. Nonetheless, the increase in DMI with current-injection in current-induced domain-wall motion highlights an interesting feature for easily tuning the DMI by current-injection.

Another study investigated the current-induced DMI in W/CoFeB/MgO systems [85], and similarly, the magnitude of DMI was quantified as a function of current density using current-induced domain wall motion. The study found an increase in DMI with increased current density, see Fig. 4 (b), but after reaching a threshold current, the DMI is reduced again. Pt/Co-based heterostructures investigated in the same study display a nearly current-independent DMI, which showcases that the physics of the current-induced DMI is not yet fully understood.

5.1 Current-induced DMI in the DFT framework

Within density-functional theory (DFT) the standard approach to compute the equilibrium symmetric and antisymmetric exchange constants is to extract them from the total energy variation associated with changes of the magnetic texture,

such as modifications of the spin spiral wave vector [86, 87, 88]. However, the application of electric current to a magnetic bilayer pushes it out of equilibrium. Since for metals out of equilibrium one cannot easily define the total energy, these recipes developed for the calculation of the equilibrium exchange constants do not apply.

An important development, which has helped researchers to formulate expressions for the exchange constants within the DFT framework that are valid also under non-equilibrium conditions, are the works that express the exchange interaction explicitly in terms of the electronic structure properties, such as the band energies, the Berry curvatures, or the single-particle Green function [89, 90, 91, 92, 93]. In the case of DMI one obtains an expression in terms of a mixed Berry curvature, which may be simplified with the help of perturbation theory into a useful approximation of DMI by the ground-state spin current [72, 73]. This result already suggests that excitation of a spin current with a suitable symmetry by an applied electric current or by a laser will change the DMI. Moreover, a study of DMI in the Rashba model using Green function techniques reports the possibility to change DMI by a gate voltage [93].

In the case of the symmetric exchange, not only the mixed Berry curvature enters the expression, but also the mixed quantum metric and the quantum metric in magnetization space, which all describe the geometrical properties of the electronic structure in mixed phase space [92]. Since spintronic effects such as the spin-transfer torque and the spin-orbit torque depend on the geometrical properties of the electronic structure as well, they are related to the exchange constants [91]. Therefore, some components of the spin-transfer torque and of the spin-orbit torque may be interpreted as a form of nonequilibrium exchange interaction [94].

The idea to change DMI by excitation of spin currents with suitable symmetry is qualitatively useful, but it is unclear if the quantitative relation [72, 73] between ground-state spin-currents and equilibrium DMI holds also under nonequilibrium conditions. Therefore, a theory has been developed to describe the induced DMI valid in the first order of the perturbation by the applied electric current [95]. Remarkably, the current-induced DMI needs to be included into the theory of current-induced torques in noncollinear magnets in order to satisfy the Onsager reciprocity relations. The mutual interrelations between various direct and inverse torques is illustrated in Fig. 5.

The strategy used to derive an expression for CIDMI in Ref. [95] makes use of Onsager reciprocity: Since the electric current driven by a time-dependent gradient in the magnetization direction is accessible by the Kubo formalism, an expression for this quantity is derived and the CIDMI is related to it by the Onsager reciprocity. This strategy to derive CIDMI from its reciprocal effect avoids completely the notion of DMI as a change of the energy due to a magnetization gradient, which is difficult to generalize for metals out of equilibrium.

CIDMI was first observed in Refs. [77, 85]. The experimental setups used in Refs. [77, 85] do not allow a CIDMI linear in the applied electric current due to symmetry reasons. Therefore, the CIDMI observed in these experiments is dominantly second order in the applied electric current. Interestingly, the

strategy used in Ref. [95] to obtain an expression for the CIDMI first order in the applied electric current cannot be generized easily to the second order: While we can compute the response of the square of an operator, e.g. the response of the square of the electric current operator, to a perturbation, e.g. the perturbation by a time-dependent gradient in the magnetization direction, the inverse is not guaranteed to provide a meaningful number. If one wants to compute the response to the applied electric current at the second order, one uses second order Kubo formalism, and not first order Kubo formalism with the square of the perturbation. Therefore, there is no obvious Onsager-relation between the CIDMI and its inverse at the second order in the applied electric current.

Different strategies are therefore required to compute the CIDMI at the second order in the applied electric current. One possible strategy is to consider the expression [91]

$$D_{ij} = \frac{1}{V}\langle r_j \mathcal{T}_i \rangle \tag{1}$$

for the DMI coefficient. Since the operator r_j is unbounded, this expression cannot be evaluated directly with Bloch-periodic wavefunctions in infinite crystals. However, in insulators the ground state charge density is simply the sum of the charge densities of all occupied Wannier functions. Therefore, it seems possible to evaluate Eq. 1 in the same way as the orbital magnetization in insulators [96, 97]. The close relation between the Berry-curvature approach to DMI and the modern theory of orbital magnetization is discussed in detail in Ref. [91]. Using Wannier functions, one may evaluate Eq. 1 also in the presence of an applied electric field and access the contribution at the second order in the perturbing electric field.

Another possible strategy to compute CIDMI at the second order in the applied electric field can be found if one interprets CIDMI as a special case of nonequilibrium exchange interactions [98]. In canted antiferromagnets, one way to assess the nonequilibrium exchange interaction is the mapping of the nonequilibrium magnetization dynamics to the Landau-Lifshitz equation [99, 98]. One may also obtain the exchange constants by considering infinitesimal rotations of spins. This method is widely applied to compute the equilibrium exchange constants. However, a generalization to the time-dependent case has been developed [100] for the multi-band Hubbard model without spin-orbit interaction. Since spin-orbit interaction has not been included, the Dzyaloshinskii-Moriya interaction is not present. However, the method has been used to assess the nonequilibrium symmetric exchange [101, 102] and one could extend it to cover nonequilibrium DMI as well.

The strategy of Ref. [100] is to derive expressions for exchange parameters in terms of non-equilibrium Green functions and self-energies by simplifying the effective action. Another approach to obtain expressions for DMI in and out of equilibrium from the effective action is described in Ref. [103]. The authors show that the ac laser-field of a laser induces a dc nonequilibrium DMI. It is likely that the same approach may be used to derive expressions for nonequilibrium DMI driven by a dc electric current. It should be noted, however, that it is not sufficient to take simply the limit $\omega \to 0$ in the ac expressions. The reason is

that a dc response from the second order perturbation by an ac electric field at optical frequencies involves a rectification effect: The phases $e^{i\omega t}$ and $e^{-i\omega t}$ are multiplied and yield a dc response. However, the second harmonic contributions $e^{2i\omega t}$ and $e^{-2i\omega t}$, which are discarded in the calculation of the dc response to optical perturbation, may contribute to the dc response when the limit $\omega \to 0$ is taken. As a consequence, the expressions for the dc photocurrent excited at optical frequencies on the one hand, and the expressions for the dc unidirectional magnetoresistance at the other hand, are not simply related by taking the limit $\omega \to 0$ [104]. A similar difference in the formulas is expected when one compares theories of dc nonlinear CIDMI and nonequilibrium DMI driven by laser pulses.

6 Summary

In conclusion, the Dzyaloshinskii-Moriya interaction (DMI) has made significant contributions to new discoveries and device concepts in the field of spintronics. Apart from its detailed study, the emergence of many chiral skyrmions in magnetic systems can be directly attributed to the presence of DMI. Multiple mechanisms exist to introduce this type of antisymmetric exchange interaction. Inversion symmetry breaking, a necessary condition for DMI, can naturally occur in the crystal lattice itself, as observed in B20 materials, which typically stabilizes chiral Bloch skyrmions. Alternatively, it can be induced by a composition gradient in thicker film-like structures. In thin films, the interfacial DMI (iDMI) induced by ferromagnetic/heavy metal interfaces has become the preferred method to introduce a significant amount of DMI. Additionally, this iDMI stabilizes Néel-type domain walls in ultra-thin films. Recently, the discovery of an interlayer DMI (IL-DMI) opens up new avenues for studying three-dimensional magnetic textures, which is an emerging field closely related to DMI. Spin structures in 3D are an emerging field of research [60], where the introduced complexity of the spin structures lends itself to advanced devices for instance in unconventional computing. Despite being discovered several decades ago, the DMI has not been fully understood, as evidenced by the recent discovery of a current-induced DMI (CIDMI). Further research is necessary to gain a comprehensive understanding of the full extent of DMI and explore potential new applications that await discovery. This may entail the exploration of three-dimensional magnetic textures that can be manipulated by CIDMI or the utilization of IL-DMI to achieve field-free switching capabilities. An intriguing avenue involves the synergistic combination of various forms of DMI. By manipulating the CIDMI, a superposition of bulk DMI (bDMI) induced by a composition gradient and iDMI could lead to an amplification or attenuation of the total DMI in the system. Such an approach has the potential to enable fine-tuning of the magnitude of the DMI in the system.

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8 Figures

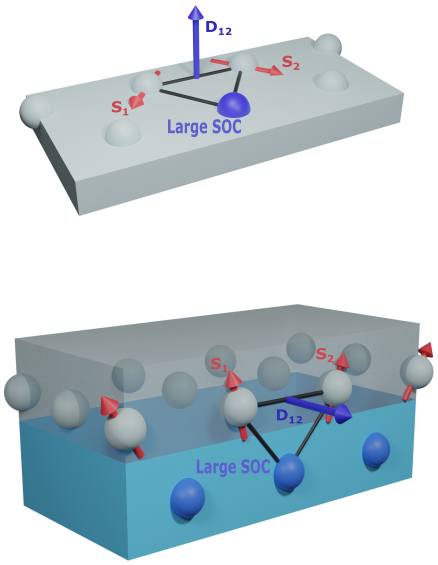

Figure 1: (Color Online) Illustration of DMI. (Top) The interaction of two magnetic atoms with an atom with large spin-orbit coupling (SOC) yields a canting of spins around the DMI vector $\boldsymbol{D}_{12}$. (Bottom) In thin films DMI is typically induced by a layer with large SOC in contact with the ferromagnetic layer.

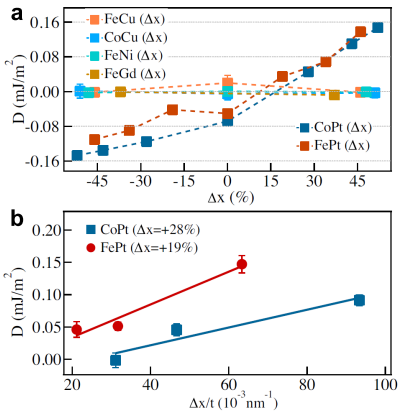

Figure 2: (Color Online) (a) Volume averaged DMI as function of change in composition Δx of a composite material A_xB_{1-x}. (b) DMI as function of change in composition Δx per nm film thickness. Reprinted with permission from Ref. [19] Copyright (2022) by the American Physical Society.

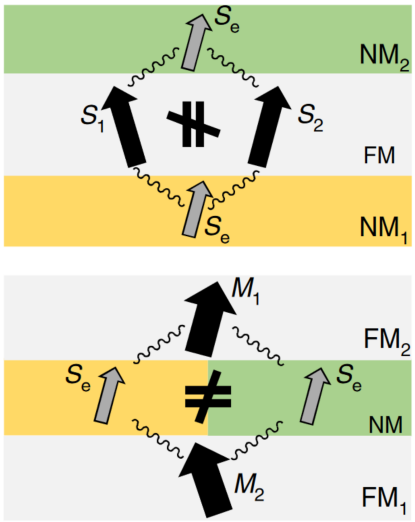

Figure 3: (Color Online) Sketch of iDMI (top) and IL-DMI (bottom). In case of iDMI the inversion symmetry breaking is from top to bottom by two different non-magnetic materials NM_1 and NM_2 sandwiching the ferromagnetic layer FM, yielding a canting of the spins S_1 and S_2 within the plane. For IL-DMI the inversion symmetry breaking is within the NM plane and yields a canting of magnetizations M_1 and M_2. Material from Ref. [65] Copyright ł 2019, The Author(s), under exclusive licence to Springer Nature Limited.

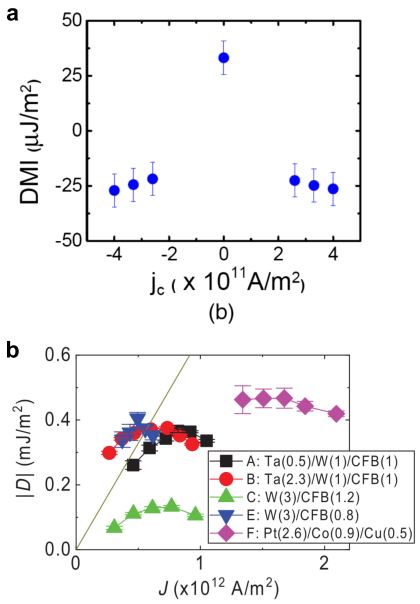

Figure 4: (Color online) (a) DMI measured in a Ta/CoFeB/MgO system via field-induced and current-induced domain wall motion.Reprinted (abstract/excerpt/figure) with permission from Ref. [77] Copyright (2018) by the American Physical Society. (b) CIDMI measured in W/CoFeB/MgO systems. Reprinted (abstract/excerpt/figure) with permission from Ref. [85] Copyright (2019) by the American Physical Society.

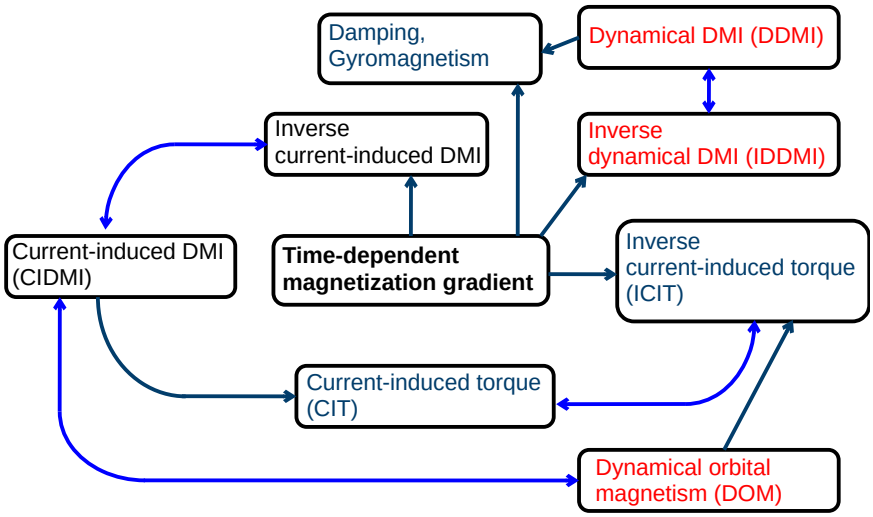

Figure 5: (Color online) Contributions driven by time dependent magnetization gradients to various torques and asymmetric exchange interactions.

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