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Bulk Sublattice Generalized Dzyaloshinskii-Moriya Interaction in PT-Symmetric Antiferromagnets

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The Dzyaloshinskii-Moriya interaction (DMI) in magnetic materials plays an important role in spintronics giving rise to chiral spin textures such as the nucleation and propagation of domain walls and skyrmions. The necessary ingredient for the emergence of the DMI is lack of inversion symmetry combined with elements with strong spin-orbit coupling (SOC). We report on a first principles investigation of generalized DMIs (gDMIs) in bulk centrosymmetric crystals with non-centrosymmetric local sublattices, where we consider a prototype family of PT-symmetric antiferromagnets, including, Mn₂Au, MnPd₂ and MnCuAs in the tetragonal and orthorhombic phases. We employ a Greens function approach to calculate the inter-atomic relativistic exchange coupling which can in turn determine the *sublattice* elements of the generalized fifth-order DMI tensor $D_{\alpha\beta,\gamma}^{ss'}$. We demonstrate that the breaking of the local mirror symmetry and the resulting local Rashba-type spin-momentum locking yields local DMIs with opposite signs on the two antiparallel sublattices. We present numerical results and provide analytical expressions for the magnon dispersion in the presence of the sublattice DMI, and show that the intra-(inter) sublattice DMIs results in identical (opposite) contributions to the nonreciprocal components of the two low-frequency AFM magnon modes.

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Antiferromagnetic (AFM) materials have attracted a lot of attention as potential candidates for active elements in spintronic devices, due to their weak sensitivity to magnetic field perturbations, potential for miniaturization, and ultrafast switching in the terahertz (THz) frequency range[1–5]. The current-induced antidamping- and field-like spin orbit torque (SOT) has been employed to control and switch the Néel vector in the two prototypical bulk centrosymmetric metallic Mn₂Au[6–8] and Dirac semimetal MnCuAs[9] antiferromagnets, thus demonstrating a viable approach to the AFM-based memories and providing a route to ultrafast spintronic devices. On the other hand, the Néel vector can be detected using the anisotropic magnetoresistance (AMR) effect[1, 4, 9], optical methods[10–12], the anomalous Hall effect in noncollinear antiferromagnets such as Mn₃X (X = Ge, Sn, Ga, Ir)[13–15] and the anisotropic spin Hall effect[16–18]. Another intriguing topological antiferromagnet is the Dirac nodal line semimetal MnPd₂ in which reorientation of the Néel vector leads to switching between the symmetry-protected degenerate state and the gapped state associated with the dispersive Dirac nodal line at the Fermi energy[18].

The AFM ordering in Mn₂Au, CuMnAs, and MnPd₂ breaks both the time-reversal symmetry, T and the space inversion symmetry, P, but preserves the PT product[6, 9, 18]. The breaking of the local space inversion on each sublattice layer can be readily seen from Fig. 1a, where, e.g., the spin-up Mn layers are sandwiched between Au and spin-down Mn layers. This results in the presence of Rashba-type spin-momentum locking with

opposite chirality[19–24] on the two magnetic Mn sublattices, which can in turn be employed to induce Néel SOT and reorient the direction of the AFM ordering.[6, 7, 25] Furthermore, *ab initio* calculations revealed the emergence of toroidal moments[26] in the Mn₂Au and CuMnAs antiferromagnets and showed that their experimentally reported SOT behaviors are consistent with the final states aligning their toroidal moments parallel to the applied electric current.[26, 27]

The Dzyaloshinskii-Moriya interaction (DMI)[28, 29] emerges in magnetic systems with artificially or intrinsically broken inversion symmetry. This relativistic antisymmetric exchange interaction results from the strong spin orbit coupling (SOC) and often favors noncollinear magnetic states, thus leading to the formation of spin spirals or topologically non-trivial spin structures such as skyrmions[30–32]. In addition, the DMI results in a non-reciprocal dispersion for long-wavelength spin-waves,[33], $\omega_{\vec{q}} = \sum_{\alpha\beta} D_{\alpha\beta} m_{\alpha} q_{\beta} \pm \omega_{\vec{q}}^0$, where, $\alpha, \beta = x, y, z$, $\omega_{\vec{q}}^0$ is an even function of the wavevector $\vec{q}$, $D_{\alpha\beta}$ represents the DMI tensor elements and $\vec{m}$ is the unit vector along the net magnetization[34, 35].

Due to the absence of a net magnetization in AFM materials, the Landau-Lifshitz-Gilbert (LLG) equation of motion describing their low-energy spin dynamics must be extended to include the sublattice dynamics of the antiparallel magnetic moments. Consequently the inter-sublattice exchange coupling, λ , between the two sublattices plays an important role in the AFM resonance (AFMR) frequency, $\omega \approx \sqrt{2K\lambda}/|M_s|$ [36, 37], resulting in a the THz range dynamics. Here, K is the magneto-crystalline anisotropy and $|M_s|$ is the amplitude of the magnetic moment of each sublattice. Moreover, even in the case of ferromagnets (FM) with two different magnetic elements, the sublattice dynamics re-

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sult in spin nutation resonance[38]. Using first principles calculations[37, 39] it was also suggested that in AFM materials, the inter-sublattice spin exchange interaction results in a significant inter-sublattice Gilbert damping that is expected to play a significant role on the AFMR linewidth of the high frequency AFMR modes.

While the DMI has been extensively studied both theoretically[40–43] and experimentally[44–49] in ferromagnetic-based bulk and interfacial systems, and in multiferroic systems[50–52] where it can give rise to weak canted ferromagnetism, there have been very few investigations of the DMI in metallic antiferromagnets[53]. Motivated by the crucial role of the intersublattice dynamics in AFM materials[37, 39] we have developed a computational approach of the interatomic relativistic exchange coupling[34, 54] which allows to determine both the inter- and intra-sublattice contributions to the DMI in bulk PT-symmetric AFM systems. Interestingly, we find that the breaking of the local mirror symmetry and the resulting local Rashba-type spin-momentum locking with opposite signs on the two sublattices yields local DMIs with opposite signs on the corresponding AFM sublattices. We investigate the effect of the sublattice DMIs on the AFM magnon dispersion and demonstrate that although the net DMI is zero, the sublattice DMIs, result in nonreciprocal magnon dispersion for both low-frequency (in-plane) and the high-frequency (out-of-plane) modes.

Theoretical Formalism: For a system with multiple sublattices, we introduce the *general* definition of the DMI contribution to the magnetic energy in continuum theory, described by the second-order expansion with respect to the local magnetization orientation and its spatial gradient [55–59], namely,

$$E_M^{gdm} = \sum_{ss'\alpha\beta\gamma} \int dV \mathcal{D}_{\alpha\beta,\gamma}^{ss'} m_\alpha^s \frac{\partial m_\beta^{s'}}{\partial R_\gamma}. \quad (1)$$

Here s, s' indicate the sublattice indices, $\alpha, \beta, \gamma = x, y, z$, the expansion coefficients, $\mathcal{D}_{\alpha\beta,\gamma}^{ss'}$, are the *generalized* DMI (gDMI) tensor elements, and $\vec{m}_s$ is the unit vector along the local magnetic moment orientation of the s th sublattice. Using integration by parts in Eq. (1) yields that only the antisymmetric DMI tensor elements, $\mathcal{D}_{\alpha\beta,\gamma}^{ss'} = -\mathcal{D}_{\beta\alpha,\gamma}^{s's}$, contribute to the DMI energy, which is often referred to as the Lifshitz invariance relation. The higher order terms in Eq.(1) in the magnetization orientation can be incorporated into an anisotropic contribution to the DMI tensor that violates the Lifshitz invariance[60].

In systems with a single sublattice, $s = s' = 1$, one can ignore the upper-indices s, s' and use the Lifshitz invariance to reduce the gDMI tensor rank to the 2nd order conventional DMI tensor (i.e. a 3×3 matrix) through, $D_{\alpha\beta} = \sum_{\gamma\gamma'} \epsilon_{\alpha\gamma\gamma'} \mathcal{D}_{\gamma\gamma',\beta} / 2$, where $\epsilon_{\alpha\gamma\gamma'}$ represents the Levi-Civita symbol.[34] However, for multi-sublattice systems it can be shown that only the symmetric components of the gDMI in sublattice space contribute to the conventional DMI, where $D_{\alpha\beta}^{ss'} = \sum_{\gamma\gamma'} \epsilon_{\alpha\gamma\gamma'} (\mathcal{D}_{\gamma\gamma',\beta}^{ss'} +$

$\mathcal{D}_{\gamma\gamma',\beta}^{s's})/4$. As we show below, the anti-symmetric inter-sublattice components of the gDMI in PT-symmetric AFMs, $(\mathcal{D}_{\alpha\beta,\gamma}^{ss'} - \mathcal{D}_{\alpha\beta,\gamma}^{s's})/2$, can potentially be nonzero, which in turn necessitates the use of gDMIs rather than the conventional DMIs.

The gDMI tensor elements are calculated using the relativistic exchange coupling, where the magnetic energy is given by,

$$E_M = -\frac{1}{2} \sum_{ij;\vec{R}\vec{R}';\alpha\beta} J_{\vec{R}-\vec{R}'}^{ij,\alpha\beta} m_\alpha^i(\vec{R}) m_\beta^j(\vec{R}'). \quad (2)$$

Here, $m_\alpha^i(\vec{R})$ is the α -component of the unit vector along the local magnetic moment of the i th atom in the unit cell at $\vec{R}$, and $J_{\vec{R}-\vec{R}'}^{ij,\alpha\beta}$ is the exchange coupling between the two local moments. The Fourier transform of the relativistic exchange coupling can be written as,

$$J_{\vec{q}}^{ss',\alpha\beta} = \sum_{i \in s, j \in s'; \vec{R}} J_{\vec{R}}^{ij,\alpha\beta} e^{i\vec{q} \cdot (\vec{R} + \vec{r}_i - \vec{r}_j)}, \quad (3)$$

where $\vec{r}_i$ and $\vec{r}_j$ are the positions of the i th and j th atoms in the unit cell and $\vec{R}$ denotes the vector connecting two different unit cells. The gDMI tensor elements are then calculated from,

$$\mathcal{D}_{\alpha\beta,\gamma}^{ss'} = \frac{1}{2V_M} \text{Im}(\partial J_{\vec{q}}^{ss',\alpha\beta} / \partial q_\gamma) |_{\vec{q}=\Gamma}, \quad (4)$$

where V_M is the unit cell volume and Γ is the center of the Brillouin Zone.

Computation Details: Structural relaxations were carried using the Vienna ab initio simulation package (VASP) [61, 62] within the generalized gradient approximation as parameterized by Perdew, Burke, and Ernzerhof (PBE).[63]. The pseudopotential and wave functions are treated within the projector-augmented wave (PAW) method [64, 65]. The plane wave cutoff energy is 500 eV and a $10 \times 10 \times 10$ k-points mesh is used in the 3D Brillouin Zone (BZ) sampling. In order to examine the effect of exchange correlation functional on the equilibrium structural and magnetic properties we have carried out calculations employing the local spin density approximation (LSDA)[66] as parameterized by Perdew and Zunger[67], the PBE[63], the LSDA+U and PBE+U[68] functionals. The PBE was found to yield the best agreement with experiment for the structural properties of all AFM systems considered in this work, and hence was used to calculate the gDMI and MCA.

The density functional theory calculations of the gDMI, magneto-crystalline anisotropy energy, and inter-sublattice exchange constant for the bulk AFM materials were carried out using the OpenMX[69–71] *ab initio* package. We adopted the Troullier-Martins type norm-conserving pseudopotentials[72] with partial core correction and the PBE exchange correlation functional[63]. We used a $24 \times 24 \times 24$ k-point mesh in the first BZ and an energy cutoff of 350 Ry for numerical integrations in real space grid.

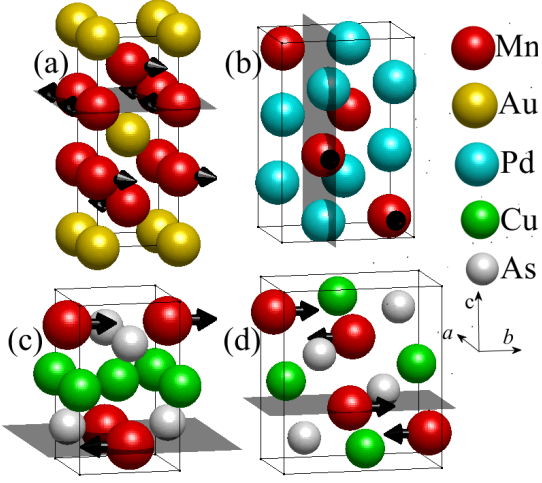


FIG. 1. Crystal structures of collinear PT-symmetric anti-ferromagnets (a) Mn_2Au (space group $I4/mmm$), (b) MnPd_2 (space group $Pnma$), (c) tetragonal MnCuAs (space group $P4/nmm$) and (d) orthorhombic MnCuAs (space group $Pnma$). The arrows denote the direction of the magnetic moments and the gray planes denote the sublattice planes which break the local mirror symmetry.

Within the linear combination of atomic orbitals scheme, the multi-orbital Hamiltonian, $\hat{H}_{\vec{R}}$, describes the hopping of electrons between unit-cells separated by distance $\vec{R}$. The electronic Hamiltonian for a magnetic system[34] in real space (which has the same form for both FM and AFM systems) can be written as,

$$\hat{\mathbf{H}} = \hat{\mathbf{H}}^0 + \hat{\mathbf{H}}^{SOC} + \sum_{i\vec{R}} \hat{\Delta}^{i\vec{R}} \vec{m}_i(\vec{R}) \cdot \vec{\sigma}, \quad (5)$$

where, the bold symbol denoted matrices in real space with elements given by, $[\hat{\mathbf{F}}]_{\vec{R},\vec{R}'} = \hat{F}_{\vec{R}-\vec{R}'}(\hat{F}$ denotes any operator in the Hamiltonian). Here, $\hat{H}_{\vec{R}}^0$ is the paramagnetic (spin-independent) term, $\hat{H}_{\vec{R}}^{SOC}$ is the SOC term and $\hat{\Delta}^{i\vec{R}}$ is the atomistic exchange splitting matrix responsible for magnetism of the i th atom. The position-dependent exchange coupling can be determined by multiplying the total exchange coupling matrix by the position operator $\hat{\Delta}^{i\vec{R}} = (\hat{1}_{i\vec{R}}\hat{\Delta} + \hat{\Delta}\hat{1}_{i\vec{R}})/2$, where $\hat{1}_{i\vec{R}}$ is the atomic position operator within the unit cell at $\vec{R}$ (diagonal matrix elements equal to one for atomic orbitals of the $(i, \vec{R})$ th atom and zero otherwise). Note that the SOC and exchange splitting terms are even and odd with respect to the time reversal symmetry, respectively, which were in turn used to decompose the total Hamiltonian into the corresponding components.

The relativistic exchange coupling matrix elements are

calculated using the Greens function method,

$$J_0^{i,zz} = \int \frac{dE}{2\pi} \text{ImTr} \left([\hat{\Delta}\hat{G} + \hat{G}\hat{\Delta}]_{\vec{R}=0} \hat{\sigma}^z \hat{1}_i \right) f(E), \quad (6a)$$

$$J_{\vec{R}}^{ij,\alpha\beta} = \frac{1}{4\pi} \int dE \text{ImTr} \left([\hat{\Delta}\hat{G}]_{\vec{R}} \hat{1}_i \hat{\sigma}^\alpha [\hat{\Delta}\hat{G}]_{-\vec{R}} \hat{\sigma}^\beta \hat{1}_j + [\hat{G}\hat{\Delta}]_{\vec{R}} \hat{1}_i \hat{\sigma}^\alpha [\hat{G}\hat{\Delta}]_{-\vec{R}} \hat{\sigma}^\beta \hat{1}_j + [\hat{G}\hat{\Delta}]_{\vec{R}} \hat{1}_i \hat{\sigma}^\alpha [\hat{\Delta}\hat{G}\hat{\Delta}]_{-\vec{R}} \hat{\sigma}^\beta \hat{1}_j \right) f(E). \quad (6b)$$

Here, $[\hat{F}\hat{G}]_{\vec{R}} = \frac{1}{N_k} \sum_{\vec{k}} \hat{F}_{\vec{k}} \hat{G}_{\vec{k}} e^{-i\vec{k}\cdot\vec{R}}$ ($\hat{F}$ and $\hat{G}$ denote any two operator in the integrand), N_k is the number of k-points, $f(E)$ is the Fermi-Dirac distribution function and

$$\hat{G}_{\vec{k}} = (E\hat{O}_{\vec{k}} - \hat{H}_{\vec{k}})^{-1}, \quad (7)$$

is the Green's function, where the Fourier transformed Hamiltonian, $\hat{H}_{\vec{k}} = \sum_{\vec{R}} \hat{H}_{\vec{R}} e^{i\vec{k}\cdot\vec{R}}$, and similarly the overlap, $\hat{O}_{\vec{k}}$, matrices are calculated using the Linear Combination of Pseudo-Atomic Orbital (LCPAO) approach as implemented in the OpenMX package.[69–71] The pseudoatomic orbitals are: Mn6.0s-3p2d1, Au7.0s-3p2d1, Cu6.0s-3p2d1, As7.0s-3p2d1 and Pd7.0s-3p2d1 (see www.openmx-square.org for details), where the abbreviation of the basis functions, such as X5.0-s1p1, X denotes the atomic element, 5.0 the cutoff radius in the generation by the confinement scheme, and s1p1 means that the calculations are carried out using one orbital for each of the s and p atomic orbitals. In the calculation of the exchange coupling the energy integration in Eq. (6a) was carried out using the Matsubara summation approach with the poles obtained from Ozaki's continued fraction method of the Fermi-Dirac distribution function.[73] The Fermi-Dirac distribution function temperature was set at $k_B T = 25$ meV and 60 poles were employed for the Matsubara summation.

The uniaxial magneto-crystalline anisotropy (MCA) constants K_u^x and K_u^y are calculated using the torque approach for single domain magnetization.[74]

Results and Discussion: We find that the total gDMI, $\mathcal{D}_{\alpha\beta,\gamma}^{tot} = \sum_{ss'} \mathcal{D}_{\alpha\beta,\gamma}^{ss'}$ vanishes for PT-symmetric materials where the global mirror symmetry is preserved. Consequently, the intra- and inter-sublattice tensor elements satisfy, $\mathcal{D}_{\alpha\beta,\gamma}^{\uparrow\uparrow} = -\mathcal{D}_{\alpha\beta,\gamma}^{\downarrow\downarrow}$, and $\mathcal{D}_{\alpha\beta,\gamma}^{\uparrow\downarrow} = -\mathcal{D}_{\alpha\beta,\gamma}^{\downarrow\uparrow}$. The numerical results of the relativistic exchange coupling reveal that the nonzero relativistic gDMI tensor elements are of the form,

$$\mathcal{D}_{\alpha\beta,\gamma}^{ss'} = -\mathcal{D}_{\beta\alpha,\gamma}^{s's} = \delta_{\alpha\gamma} \vec{D}_g^{ss'} \cdot (\vec{e}_\beta \times \vec{e}_\alpha). \quad (8)$$

Here, $\vec{D}_g^{ss'}$ are the sublattice matrix elements of the gDMI vector, $\delta_{\alpha\gamma}$ is the Kronecker delta, and $\vec{e}_{\alpha(\beta)}$ is the unit vector along the $\alpha(\beta)$ direction. Throughout the remaining manuscript we denote the anti-symmetric inter-sublattice gDMI with $\vec{D}_{g,as}^{\uparrow\downarrow}$. We also find that the Lifshitz invariance identity, $\mathcal{D}_{\alpha\beta,\alpha}^{ss'} = -\mathcal{D}_{\beta\alpha,\alpha}^{s's}$ is satisfied

in the numerical calculations. Note that, in the case of Néel type DMIs the unit vector $\vec{e}_\beta$ points along the broken local mirror symmetry direction. Therefore, for an AFM with broken local mirror symmetry along, e.g., the z -axis, the gDMI vector $\vec{D}_g^{ss'}$ lies in the xy -plane.

In addition to the fact that $\vec{D}_{g,as}^{\uparrow\downarrow}$ does not contribute to the conventional DMI, $D_{\alpha\beta}^{ss'}$, it can also be shown that the anti-symmetric inter-sublattice gDMI elements do not originate from the anti-symmetric inter-atomic exchange coupling (i.e. inter-atomic DMI), where the magnetic energy can be written as $E_{DMI} = \sum_{ij} \vec{d}_{ij} \cdot \vec{m}_i \times \vec{m}_j$ (i, j denote atomic sites). In this case, the corresponding contribution to the relativistic exchange coupling for a system with broken mirror symmetry along the z -axis is given by, $J_{ami}^{ij,\alpha\beta} = \sum_\gamma \epsilon_{\alpha\beta\gamma} d_{ij}^\gamma$, where $\vec{d}_{ij} = d_{r_{ij}}^N \vec{e}_z \times \vec{r}_{ij}/r_{ij}$, and $d_{r_{ij}}^N$ is the atomistic Néel DMI strength between two atoms separated by $\vec{r}_{ij}$. The presence of the Levi-Civita symbol in the relativistic exchange coupling results in gDMI tensor elements that obey, $\mathcal{D}_{\alpha\beta,\gamma}^{ss'} = -\mathcal{D}_{\beta\alpha,\gamma}^{ss'}$, leading to a *symmetric* contribution to the inter-sublattice gDMI vector, $\vec{D}_g^{\uparrow\downarrow} = \vec{D}_g^{\downarrow\uparrow}$, which is absent in Eq. (8). This suggests that the anti-symmetric gDMI term in Eq. (8) originates from the off-diagonal components of the anisotropic inter-atomic exchange coupling and therefore should not be confused with the conventional inter-sublattice DMI present in systems with broken global mirror symmetry. The unconventional nature of the inter-sublattice gDMI is further demonstrated below where we find that it varies quadratically with the SOC, in sharp contrast to the linear SOC dependence of the conventional DMI.[28]

In addition to the nonzero matrix elements in Eq. (8), we find finite values for $\mathcal{D}_{\alpha\alpha,\beta}^{ss}$, which are of non-relativistic origin and arise from the variation of the intersublattice exchange coupling between successive Mn layers along the locally broken mirror symmetry direction. This can be understood using a 1d magnetic chain along the z -axis with two magnetic moments within the unit-cell at the origin and the relative position, b/a , where a is the length of the unit cell. Considering staggered intra- and inter-cell exchange interactions, J_1 and J_2 , respectively, and using Eqs. (3) and Eq. (4) we find $J_{q_z}^{12,zz} = (J_{q_z}^{21,zz})^* = e^{-iq_z b}(J_1 + J_2 e^{iq_z a})$ and $D_{zz,z}^{12} \propto (a-b)J_1 - bJ_2$. It should be noted that the last term appear in the off-diagonal elements of the magnon Hamiltonian and does not yield a nonreciprocal dispersion to the magnon band structure.

Table. I lists the calculated values of the relaxed lattice constants, magnetic moment, uniaxial MCA, and sublattice gDMI tensor elements, $\vec{D}_g^{\uparrow\uparrow}$ and $\vec{D}_{g,as}^{\uparrow\downarrow}$ for Mn_2Au (space group I4/mmm), MnPd_2 (space group Pnma), tetragonal MnCuAs (space group P4/nmm) and orthorhombic MnCuAs (space group Pnma), respectively. The corresponding broken local mirror symmetry planes of the crystal structures are shown in Figs. 1(a)-(d), respectively. The numbers in parenthesis denote the corre-

sponding experimentally reported values. Overall there is good agreement for the lattice constants and the magnetic moments. The negative MCAs suggest in-plane magnetic anisotropy for all four compounds, which are also in agreement with the experimental observations. Interestingly for Mn_2Au , $\vec{D}_g^{\uparrow\uparrow} \approx \vec{D}_{g,as}^{\uparrow\downarrow}$, while for the remaining AFMs the inter-sublattice gDMI is much smaller compared to the intra-sublattice DMI.

The gDMI vectors for Mn_2Au and tetragonal MnCuAs are isotropic in the xy -plane due to their tetragonal crystal structure, while for orthorhombic MnCuAs the intra-sublattice gDMIs are slightly anisotropic. On the other hand, for MnPd_2 the gDMI vector lies on the xz -plane because the local mirror symmetry is broken along the y -axis, with the x - and z - components having opposite signs.

In order to understand the underlying mechanism for the sublattice-resolved gDMI in Mn_2Au , where $\vec{D}_g^{\uparrow\uparrow} \approx \vec{D}_{g,as}^{\uparrow\downarrow}$, we show in Figs. 2(a) and (b) the x -component of the intra-sublattice gDMI on the $\text{Mn}_\uparrow$ sublattice and the inter-sublattice gDMI, respectively, as a function of chemical potential shift. Turning on the SOC only for the $\text{Mn}_\downarrow$ (Au) layers results in negative (positive) contribution to the $\vec{D}_g^{\uparrow\uparrow} \cdot \hat{e}_x$ on the $\text{Mn}_\uparrow$ -sublattice, while the SOC of the $\text{Mn}_\uparrow$ layer yields a relatively negligible contribution. The opposite sign of the $\text{Mn}_\downarrow$ (dashed black curve) and Au (red curve) layers contributions reduce in turn the total $\vec{D}_g^{\uparrow\uparrow} \cdot \hat{e}_x$. We find that the contribution of the $\text{Mn}_\downarrow$ layer is relatively insensitive to chemical potential while that of the Au layer increases with decreasing chemical potential due to the depletion of electrons from the fully occupied Au d -orbitals resulting in sign reversal of the total $\vec{D}_g^{\uparrow\uparrow} \cdot \hat{e}_x$ with the hole-doping. On the other hand, the variation of $\vec{D}_{g,as}^{\uparrow\downarrow} \cdot \hat{e}_x$ with chemical potential in Fig. 2(b) shows the inter-sublattice gDMI originates primarily from the Au atoms since the contributions from the two Mn sublattices cancel out. Note that the relatively small difference between the total gDMI and the sum of the atom-resolved contributions is due to the fact that the atomic decomposition is accurate only to first order perturbation in SOC.

In Figs. 2(c) and (d) we present the results for the intra- and inter-sublattice gDMIs versus the SOC scaling factor which multiplies the second term of the Hamiltonian in Eq. (5). We find that, as expected from an observable that originates from the broken local mirror symmetry and local Rashba-type spin-momentum locking, the intra-sublattice gDMI is to the lowest order a linear function of SOC. In sharp contrast, the inter-sublattice gDMI, exhibits an even dependence on the SOC which is uncharacteristic of the DMI and resembles the quadratic SOC dependence of the anisotropic exchange coupling. The absence of a conventional contribution to the inter-sublattice gDMI can be understood by the fact that the two sublattices are connected through a local mirror symmetry operation.

Magnon Dispersion: In the absence of an external mag-

TABLE I. Equilibrium lattice constants, a, b, c , magnetic moment of Mn atom, $|M_{Mn}|$, uniaxial magnetocrystalline anisotropy energies, K_u^x and K_u^y , per formula unit, and intra-sublattice, $\vec{D}_g^{\uparrow\uparrow} = -\vec{D}_g^{\downarrow\downarrow}$, and anti-symmetric inter-sublattice, $\vec{D}_{g,as}^{\uparrow\downarrow}$, gDMI vectors for Mn_2Au , MnPd_2 , tetragonal MnCuAs and orthorhombic MnCuAs and respectively. The numbers in parentheses denote the experimentally reported values.

	a (Å)	b (Å)	c (Å)	$ M_{Mn} $ (μ_B)	$K_u^{x,y}$ ($\frac{\text{meV}}{\text{f.u.}}$)	$\vec{D}_g^{\uparrow\uparrow}$ (mJ/m ²)	$\vec{D}_{g,as}^{\uparrow\downarrow}$ ($\mu\text{J}/\text{m}^2$)
Mn_2Au	3.28 (3.32) ^a	3.28 (3.32) ^a	8.43 (8.53) ^a	3.5	-2.5, -2.5	(0.11, 0.11, 0)	(130, 130, 0)
MnPd_2	3.99 (4.03) ^d	5.49 (5.46) ^d	8.28 (8.13) ^d	3.88	-0.08, 0.05	(-0.11, 0, 0.2)	(17, 0, -20)
Tetragonal- MnCuAs	3.68 (3.8) ^b	3.68 (3.8) ^b	6.4 (6.3) ^b	3.4	-0.17, -0.17	(-1.13, -1.13, 0)	(-10, -10, 0)
Orthorhombic- MnCuAs	3.8 (3.86) ^c	6.45 (6.58) ^c	7.29 (7.3) ^c	3.6	-0.08, -0.11	(-0.41, -0.44, 0)	(8, 3, 0)

^a Ref.[75]; ^bRef.[76]; ^cRef.[77]; ^dRef.[78]

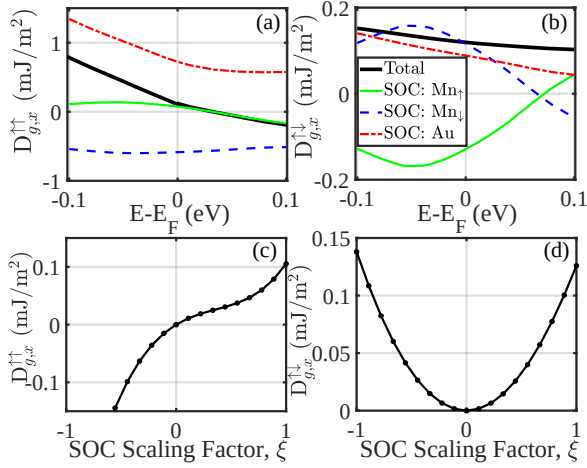


FIG. 2. Intra-sublattice (a) and inter-sublattice (b) bulk gDMI in Mn_2Au versus shift of the chemical potential. The intra-sublattice gDMI correspond to the spin-up Mn atoms. The green blue and red curves denote the layer-resolved DMIs when only the SOC of the spin-up Mn, spin-down Mn and Au atoms are turned on, respectively. Intra-sublattice (c) and inter-sublattice (d) gDMIs in Mn_2Au versus the SOC scaling factor.

netic field, the atomistic spin dynamics are governed by the LLG equation of motion for magnetization dynamics around the equilibrium configuration, $\vec{m}_0^s(\vec{R}) = m_z^s \vec{e}_z$,

$$\frac{M_s}{2} \frac{\partial \vec{m}^s(\vec{R})}{\partial t} = -\vec{m}^s(\vec{R}) \times \vec{B}_{eff}^{\vec{R},s}, \quad (9)$$

where, $m_z^s = \pm 1$ and the effective magnetic field experienced by the sublattice s at position $\vec{R}$, is given by,

$$\vec{B}_{eff}^{\vec{R},s} = J_0^{s,zz} \vec{e}_z + \sum_{s'\vec{R}'} J_{\vec{R}-\vec{R}'}^{ss',\alpha\beta} m_{\alpha}^{s'}(\vec{R}') \vec{e}_{\beta}, \quad (10)$$

with, $\alpha, \beta = x, y$. The magnon dispersion can be obtained by solving the linearized LLG equation around the equilibrium magnetization direction, m_z^s , and Fourier

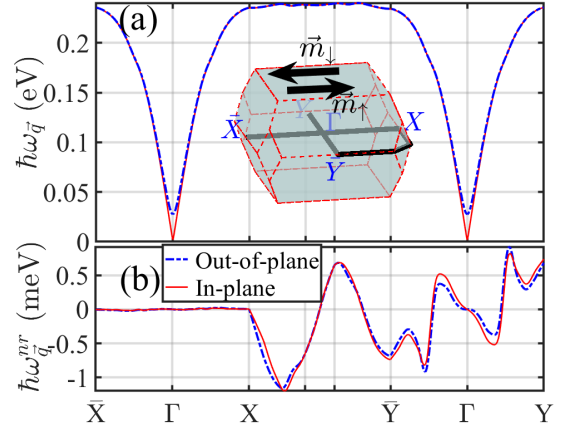


FIG. 3. (a) Magnon dispersion for Mn_2Au along high symmetry directions with Néel ordering vector along x ; (b) corresponding nonreciprocal component, $\omega_{\vec{q}}^{nr} = (\omega_{\vec{q}} - \omega_{-\vec{q}})/2$, versus high symmetry directions. The solid blue (dash-dotted red) curves correspond to the in-plane (out-of-plane) mode. Inset: Brillouin zone and the corresponding high symmetry directions.

transforming both in time and space,

$$\frac{M_s}{2} i\omega m_{x,\vec{q}}^s = m_z^s \sum_{s'\alpha} J_{\vec{q}}^{ss',\alpha y} m_{\alpha,\vec{q}}^{s'} - m_{y,\vec{q}}^s J_0^{s,zz}, \quad (11a)$$

$$\frac{M_s}{2} i\omega m_{y,\vec{q}}^s = m_{x,\vec{q}}^s J_0^{s,zz} - m_z^s \sum_{s'\alpha} J_{\vec{q}}^{ss',\alpha x} m_{\alpha,\vec{q}}^{s'}. \quad (11b)$$

It should be noted that in order to observe effects of a Néel type DMI on the magnon dispersion, the direction of the equilibrium magnetic moments should be perpendicular to the direction of broken local mirror symmetry. A generalization of the above to an arbitrary orientation for the equilibrium magnetization direction is straightforward.

The magnon band structure, $\omega_{\vec{q}}$, along the high-symmetry directions for Mn_2Au with Néel ordering pointed along the x -axis is displayed in Fig. 3(a), with the corresponding nonreciprocal component, $\omega_{\vec{q}}^{nr} = (\omega_{\vec{q}} - \omega_{-\vec{q}})/2$ shown in Fig. 3(b). The dashed-blue (solid-

red) dispersion curves correspond to the out-of-plane (in-plane) mode. Similar to the ferromagnetic case, the slope of the nonreciprocal magnon dispersion at the Γ -point yields the DMI. A nonzero value for the nonreciprocal magnon dispersion can be seen for magnons propagating along the Y-direction which is perpendicular to both the Néel ordering orientation and the broken local mirror symmetry axis (i.e. z -axis).

The different slopes of the two modes at the Γ can be understood using the analytical expression for the magnon dispersion which can be obtained by the eigenfrequencies of the 4×4 matrix given by Eqs. (11a) and (11b), which are of the form,

$$\frac{1}{2}|M_s|\omega_{\vec{q}}^{\pm} = \text{Im}(J_{\vec{q}}^{\uparrow\uparrow,xy}) \pm \text{Im}(J_{\vec{q},as}^{\uparrow\downarrow,xy}) + \omega_{\vec{q}}^{0,\pm}, \quad (12)$$

where, in the derivation we used, $J_{\vec{q},as}^{\downarrow\uparrow,xy} = -J_{\vec{q},as}^{\uparrow\downarrow,xy}$, $J_{\vec{q}}^{\downarrow\downarrow,xy} = -J_{\vec{q}}^{\uparrow\uparrow,xy}$, and,

$$(\omega_{\vec{q}}^{0,\pm})^2 = \left(J_{\vec{q}}^{\uparrow\uparrow,xx} - J_0^{\uparrow,zz} \mp J_{\vec{q}}^{\uparrow\downarrow,xx} \right) \times \left(J_{\vec{q}}^{\uparrow\uparrow,yy} - J_0^{\uparrow,zz} \pm J_{\vec{q}}^{\uparrow\downarrow,yy} \right), \quad (13)$$

is an even function of $\vec{q}$ and at $\vec{q} = \Gamma$ yields the Kittel's anti-ferromagnet resonance (AFMR) expression[36, 37]. The identical contribution of the intra-sublattice gDMI, $\text{Im}(J_{\vec{q}}^{\uparrow\uparrow,xy})$, to both magnon modes can be readily seen in the limit of independent sublattice dynamics, whereby, the two sublattices and the two corresponding magnon modes are assumed completely independent with the nonreciprocal components given by, $\omega_{\vec{q}}^{s,nr} = \sum_{\alpha\beta} D_{\alpha\beta}^{ss} m_{\alpha}^s q_{\beta}$. [34] Since the magnetic moments of the two sublattices are anti-parallel, $m_{\alpha}^{\uparrow} = -m_{\alpha}^{\downarrow}$, and the intra-sublattice gDMI tensor elements have opposite signs, $D_{\alpha\beta}^{\uparrow\uparrow} = -D_{\alpha\beta}^{\downarrow\downarrow}$, we obtain, $\omega_{\vec{q}}^{\uparrow,nr} = \omega_{\vec{q}}^{\downarrow,nr}$. This is in contrast to the effect of the total DMI, $(D_{\alpha\beta}^{\uparrow\uparrow} + D_{\alpha\beta}^{\downarrow\downarrow})/2$, which is nonzero in systems with broken global mirror symmetry and results in the shift of the two magnons in opposite directions and their hybridization at the Γ -point.[79] It is also worth noting that in the absence of MCA where the dispersion is given by $\omega_{\vec{q}}^0 \propto |\vec{q}|$, the

nonreciprocal component of the magnon dispersion, *e.g.*, $\omega_{\vec{q}}^{nr} \propto q_x$, resulting from the alternating intra-sublattice DMI, $D^{\uparrow\uparrow} = -D^{\downarrow\downarrow}$, does not shift the position of the minima of the magnon dispersion at the Γ -point but instead yields asymmetric change of the slopes of the magnon dispersion propagating along opposite directions, resulting in an antisymmetric V-shaped dispersion.

In summary, a first principles study based on the Greens function approach is presented for the DMI of a family of AFM materials with broken local mirror symmetry, including Mn_2Au , MnCuAs and MnPd_2 , which allows the separation of the intra- and inter-sublattice contributions to the generalized DMI. For the prototype Mn_2Au we elucidate the atomistic origin of the sublattice gDMI by turning on the SOC on specific atoms. We demonstrate that the intra-sublattice gDMI of the $\text{Mn}_{\uparrow}$ sublattice arises from the interplay of the Au- and spin-down $\text{Mn}_{\downarrow}$ atom-resolved contributions which have opposite sign for a wide range of chemical potential shift. On the other hand, the source of the inter-sublattice gDMI is due solely to the Au atoms. Furthermore, we show that the intra-sublattice gDMI, which is the dominant component, has a linear dependence on SOC and contributes equally to both in-plane and out-of-plane magnon modes. In contrast, the inter-sublattice gDMI tensor elements depend quadratically on the SOC and, although, it shares the same microscopic origin as the anisotropic exchange coupling, it has a nonreciprocal contribution to the magnon dispersion. Our theoretical predictions of the difference in nonreciprocal magnon dispersion for the two modes can be used to measure the sublattice components of the gDMI tensor elements.

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