# Tuning magnetic exchange interactions in two-dimensional magnets: The case of $CrGeX_3$ (X = Se, Te) and Janus $Cr_2Ge_2(Se,Te)_3$ monolayers

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We present a computational study to explore the potential of different experimental approaches to tune the magnetic interactions in two-dimensional van der Waals magnets. We selected  $CrGeSe_3$ ,  $CrGeTe_3$ , and Janus  $Cr_2Ge_2(Se, Te)_3$  monolayers as case studies and calculated the full exchange tensors among all relevant atomic pairs and analyze their dependence on different external parameters, such as biaxial and uniaxial strain, as well as gate voltage. We focus particularly on interactions that emerge or vanish due to changes in the system's symmetry, especially under uniaxial strain and gate voltage. We find that biaxial and uniaxial strains significantly modify isotropic exchange couplings, which can lead to a transition from a ferromagnetic to an antiferromagnetic phase, while a gate voltage induces Dzyaloshinskii–Moriya interactions, forming a vortex pattern whose chirality is determined by the sign of the electric field. The electric dipole moment of the Janus material is large, raising the possibility of multiferroic behavior. The polarizability is similar for the three compounds.

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# I. INTRODUCTION

Since the initial experimental report of magnetism in twodimensional (2D) materials in 2017 [1,2], the interest of the scientific community in van der Waals (vdW) magnets has greatly increased. On the one hand, they promise to be strong candidates for future magnetic memory technologies like magnetic random-access memory (MRAM)-based devices [3,4] and, due to their layered exfoliation behavior, they can successfully facilitate the miniaturization process. On the other hand, the magnetic domain structure found in 2D vdW magnets [5,6] might open the way toward developing domainwall memory technology in 2D materials [7].

Long-range magnetic order appears in 2D vdW magnets because the magnetic anisotropies arising from spin-orbit coupling (SOC) are large enough to overcome thermal and quantum fluctuations [8]. However, the experimental Curie temperatures ( $T_C$ ) of these materials remain relatively low: 35 K for CrI<sub>3</sub> monolayer [1] or 175 K for 10-layer-thick Fe<sub>3</sub>GeTe<sub>2</sub> [9]. More promisingly, SOC can lead to noncollinear magnetic configurations, such as helical states [10,11] or spin textures [12,13] that have been suggested to be candidates for data storage [14]. In most cases, these are caused by the antisymmetric exchange, i.e., the Dzyaloshinskii-Moriya (DM) interaction. In centrosymmetric materials like NiI<sub>2</sub> monolayers, where the DM interaction vanishes because of inversion symmetry, skyrmionic structures can be stabilized with the aid of symmetric exchange [15]. Other magnets that have been proposed to host noncollinear magnetic textures are Janus materials. The interest in Janus magnets lies in the fact that due to the lack of inversion symmetry it is easier to induce noncollinear magnetic textures triggered by DM interactions [16], providing a solid platform for skyrmion-based devices in the near future. Examples of Janus magnets that have been studied previously include  $Cr_2(I, X)_3$  (X = Br, Cl, F) [17,18], Ni(X,Y) (X,Y = I, Br, Cl) [19], MnBi<sub>2</sub> $X_2$ Te<sub>2</sub> (X = Se, S) [20], or Cr(Y,X) (Y = S, Se, Te; X = Cl, Br, I) [21]. To date, the synthesis of Janus monolayers has been limited, with only a few examples, such as SPtSe [22] and MoSeS [23].

The recently developed computational tool GROGU [24,25] enables us to quantify the bilinear tensorial exchange interactions between any two atomic magnetic entities from first-principles calculations, well beyond the capability of methods based on energy differences [26]. In this paper, we take advantage of this tool to analyze how different parameters, which can be controlled experimentally, affect these interactions and could even lead to a change in the magnetic configuration of a 2D vdW magnet. In particular, we consider two types of effects. One is the change in the bond distances and angles between magnetic atoms, which can be controlled by the application of strain. As we show here, strain can have a strong effect on the magnitude and even the sign of the magnetic interactions, eventually inducing a phase transition



FIG. 1. Top and side views of (a) the CGS and (b) the Janus CGST structures. Cr, Ge, Se, and Te atoms are depicted in red, blue, yellow, and grey, respectively. Lattice vectors  $\vec{a}_1$  and  $\vec{a}_2$  are shown in (a). Labeling of the nearest Cr neighbors with respect to a reference atom *c* is also shown in (b). Cr-X-Cr (X = Se or Te) bond angles are shown in (c).

between ferromagnetic (FM) and antiferromagnetic (AFM) states. A second approach consists of changing the system's symmetry, which can be achieved by atomic changes in the structure, uniaxial strain, or the application of a gate voltage. Certain magnetic interactions are forbidden in high-symmetry structures, while lowering the symmetry of the system can allow some of those interactions to appear. We show that large DM interactions can be activated and tuned by uniaxial strain, and especially by a gate voltage. Such effects then can promote the appearance of noncollinear magnetic structures with a simple experimental knob. In fact, experiments proved that a gate voltage can tune the chirality of skyrmions in centrosymmetric monolayers such as CrGeTe<sub>3</sub> (CGT) [27].

To illustrate how magnetic transitions can be triggered, we study the monolayers of CrGeSe<sub>3</sub> (CGS) and CGT. The magnetic, electronic, and magnonic properties of these vdW materials have been widely studied [28–33], and interest in them has increased in part because of the new 2D physics they can offer. CGS and CGT monolayers possess a rhombohedral structure and are characterized by the space group  $R\bar{3}$ . Their minimal unit cell contains ten atoms and the structure is depicted in Fig. 1(a). We also study a Janus version of this structure, where the top external layer is formed by Te and the bottom external by Se atoms as depicted in Fig. 1(b). This system corresponds to the Cr<sub>2</sub>Ge<sub>2</sub>(Se, Te)<sub>3</sub> (CGST) stoichiometry. Phonon spectra calculations have shown that these lattice structures are stable [16,34]. Recently, Monte Carlo calculations have shown that Janus CGST can stabilize skyrmionic states [16].

This paper aims to provide a comprehensive understanding of how symmetry breaking through external perturbations affects the exchange parameters, serving as a clear guide for future control and tuning of the magnetic properties of 2D magnets. In Sec. II, we define the parts of the spin Hamiltonian used to model the magnetic interactions and determine which ones vanish due to the symmetry of the systems under consideration. In Sec. III, we give the details of the calculations. In Sec. IV, we present and explain the results obtained when biaxial and uniaxial strain as well as gate voltage are applied. Finally, in Sec. V we summarize and draw our conclusions.

## **II. SPIN MODEL AND SYMMETRY CONSTRAINTS**

Most magnetic materials with localized magnetic moments can be described by the classical Heisenberg Hamiltonian,

$$H(\{\mathbf{e}_i\}) = \frac{1}{2} \sum_{i \neq j} \mathbf{e}_i \,\mathcal{J}_{ij} \,\mathbf{e}_j + \sum_i \mathbf{e}_i \,\mathcal{K}_i \,\mathbf{e}_i, \qquad (1)$$

where  $\mathbf{e}_i$  is a unit vector pointing along the spin-only magnetic momentum of the magnetic atom located on site *i*,  $\mathcal{K}_i$  is



FIG. 2. (a) Schematic picture of the local frame of reference, where the bond connecting the *A* and *B* atoms is parallel to the *x*-axis. The positive *z* direction points towards the reader. Symmetries and selection rules are computed with respect to the center *C*. (b)–(d) Sketch of noncollinear textures induced by DM vectors at first nearest neighbors relative to the central atom. Magnetic moments are represented by blue arrows. (b) A vortex pattern is formed around the central atom if only a radial DM component is present. (c) A radial vortex pattern is formed around the central atom if only a nagular DM component is present. In panels (b) and (c), the spin in the central atom is pointing towards the reader. (d) An in-plane relative  $90^{\circ}$ -degree orientation among first neighbors is boosted when the nearest-neighbor DM vectors are normal to the *xy* plane. The thin curved arrows demonstrate that this magnetic pattern can be rotated in the plane at zero energy cost.

the on-site anisotropy matrix on site *i*, and  $\mathcal{J}_{ij}$  is the exchange tensor that can be written as the sum of a symmetric and an antisymmetric part:

$$\begin{aligned} \mathcal{J}_{ij} &= \mathcal{J}_{ij}^{s} + \mathcal{J}_{ij}^{a} \\ &= \begin{pmatrix} J_{ij}^{xx} & S_{ij}^{z} & S_{ij}^{y} \\ S_{ij}^{z} & J_{ij}^{yy} & S_{ij}^{x} \\ S_{ij}^{y} & S_{ij}^{x} & J_{ij}^{zz} \end{pmatrix} + \begin{pmatrix} 0 & D_{ij}^{z} & -D_{ij}^{y} \\ -D_{ij}^{z} & 0 & D_{ij}^{x} \\ D_{ij}^{y} & -D_{ij}^{x} & 0 \end{pmatrix}. \end{aligned}$$
(2)

The DM vectors are then defined as  $\mathbf{D}_{ij} = (D_{ij}^x, D_{ij}^y, D_{ij}^z)$ . It is common to write the spin-Hamiltonian (1) in the following form:

$$H(\{\mathbf{e}_i\}) = \frac{1}{2} \sum_{i \neq j} J_{ij}^H \mathbf{e}_i \cdot \mathbf{e}_j + \frac{1}{2} \sum_{i \neq j} \mathbf{e}_i \mathcal{J}_{ij}^S \mathbf{e}_j + \frac{1}{2} \sum_{i \neq j} \mathbf{D}_{ij} \cdot (\mathbf{e}_i \times \mathbf{e}_j) + \sum_i \mathbf{e}_i \mathcal{K}_i \mathbf{e}_i, \quad (3)$$

where  $J_{ij}^{H} = \frac{1}{3} \text{Tr} J_{ij}^{s}$  is the isotropic Heisenberg coupling and  $\mathcal{J}_{ij}^{S} = \mathcal{J}_{ij}^{s} - J_{ij}^{H} \mathcal{I}$  is a traceless symmetric matrix.

The knowledge of the exchange constants is crucial for computing and predicting the order-disorder transition temperature  $T_c$ , the magnon spectra or the spin pair correlation functions. Moreover, external control over these constants through electric fields or strain can result in magnetic phase transitions [35–38], generate skyrmionic textures [39,40], or tune  $T_c$  [41].

In our calculations,  $\mathcal{J}_{ij}$  is computed at the local frame of reference shown in Fig. 2(a), while the corresponding atomic

TABLE I. Symmetries and nonzero exchange parameters for the first five nearest neighbors in CGS and CGST structures following the local frame of reference depicted in Fig. 2(a). For simplicity, we removed  $J^{xx}$ ,  $J^{yy}$ , and  $J^{zz}$  from the list of nonzero exchange parameters as there are no selection rules for these matrix elements.

Bond	Symmetries centered on $\ensuremath{\mathcal{C}}$	Nonzero exchange parameters
c1	$\Pi, \sigma_{yz}, C_{2x}/\sigma_{yz}$	$S^x/D^y, D^z, S^x$
c2	$C_{2y}/\emptyset$	$D^x$ , $D^z$ , $S^y/D^x$ , $D^y$ , $D^z$ , $S^x$ , $S^y$ , $S^z$
c3	$\Pi, \sigma_{yz}, C_{2x}/\sigma_{yz}$	$S^x/D^y, D^z, S^x$
c4	Π/Ø	$S^x$ , $S^y$ , $S^z/D^x$ , $D^y$ , $D^z$ , $S^x$ , $S^y$ , $S^z$
c5	$C_{2x}/\emptyset$	$D^x$ , $S^x/D^x$ , $D^y$ , $D^z$ , $S^x$ , $S^y$ , $S^z$

pairs ij are labeled as indicated in Fig. 1(b). In this figure, each pair  $ij \in \{c1, c2, c3, c4\}$  exhibits a different point group symmetry, that leads to different selection rules for the exchange constants. In the Appendix, we summarize the selection rules for the exchange constants when inversion symmetry  $(\Pi)$ , mirror  $\alpha\beta$ -plane symmetry ( $\sigma_{\alpha\beta}$ ), and  $C_2$  symmetry around the  $\alpha$  axis ( $C_{2\alpha}$ ) (where  $\alpha$ ,  $\beta = x$ , y, and z) are present with respect to the center of a given bond. We apply these rules to identify the nonzero exchange parameters from the corresponding symmetries for the CGS and CGST structures; see Table I. The Janus structure of CGST breaks  $\Pi$ ,  $\sigma_{xy}$ ,  $C_{2x}$ , and  $C_{2v}$  symmetries that are present in CGS and CGT. For CGST, the first and third neighbors have nonzero  $D^{y}$  and  $D^{z}$  values, which vanish for CGS and CGT. We will demonstrate that it is also possible to induce  $D^{y}$  in CGS and CGT between first neighbors by a gate voltage that breaks inversion symmetry. Moreover, uniaxial strain breaks  $C_{2y}$  symmetry with respect to the c2' bond, inducing a  $D_{c2'}^{y}$  component. In Figs. 2(b)–2(d), we show a sketch of how different DM vector components induce noncollinear magnetic configurations.

#### **III. COMPUTATIONAL METHOD**

We compute all terms in the spin-Hamiltonian given in Eq. (3) by a two-step process. First, we obtain the electronic Hamiltonian based on Density Functional Theory (DFT) [42,43] by using the SIESTA code [44]. This code has the advantage that electronic wave functions are expanded in a basis of pseudo-atomic orbitals, from which it is easy to identify the atomic magnetic moments. In as second step, the DFT Hamiltonian is used as input of the package GROGU [24], which extracts the exchange and anisotropy constants of the system based on the magnetic force theorem [45,46].

Our DFT calculations employed the generalized gradient approximation (GGA) in the scheme of Perdew-Burke-Ernzerhof (PBE) [47] for the exchange-correlation potential. A  $10 \times 10 \times 1$  Monkhorst-Pack *k*-point grid and a mesh cutoff of 1000 Ry for reciprocal (real-space) integrals were used for an accurate self-consistent convergence. We employed a double- $\zeta$  polarized basis set whose first- $\zeta$  PAOs had long radii of about 8 Bohr. All calculations were carried out using structural relaxations with a maximum force tolerance equal to 0.005 eV/Å. Interactions with the core electrons were included by a Troullier-Martins pseudopotential [48], obtained as described in Ref. [49]. For the GROGU calculations, a  $100 \times 100 \times 1$  Monkhorst-Pack *k*-grid was used and

TABLE II. Summary of the structural, electronic and magnetic properties of CGS, CGT and CGST obtained from the SIESTA calculations. Orbital and spin moments where computed by GROGU.

	CGS	CGT	CGST
Lattice constant (Å)	6.44	6.98	6.71
Spin moment $(\mu_B)$	3.531	3.749	3.643
Orbital moment $(\mu_B)$	0.008	0.022	0.0046
MAE (meV/Cr)	-0.05	0.80	0.15
Band gap (eV)	0.8	0.2	0.3

localized magnetic moments were projected onto the Cr 3*d*-orbitals to avoid undesired contributions from nonmagnetic orbitals [24].

#### **IV. RESULTS**

First, we obtained the structural and electronic ground state of the three systems, CGS, CGT and CGST. A summary of the results is given in Table II. The lattice constant of CGST lies in between those of CGS and CGT, as expected. As of the magnetic structure, all these systems are ferromagnetic with a similar magnetic moment localized on each Cr atom, while all other atoms are non-magnetic. The orbital contribution to the magnetic moments is below 1% an all cases. We define the magnetic anisotropy energy (MAE) as the difference of the self-consistently calculated total energy between in-plane and out-of-plane magnetizations,  $MAE = E_{\parallel} - E_{\perp}$ . Two-site  $\frac{1}{2} \sum_{i \neq j} (J_{ij}^{xx} - J_{ij}^{zz})$  and on-site  $\sum_i (K_i^{xx} - K_i^{zz})$  magnetic anisotropy contributions to the MAE can be obtained by choosing collinear in-plane (along the x axis) and out-of-plane (along the z axis) magnetizations in Hamiltonian 3. So the MAE is characterized by examining the  $J_{cj}^{xx} - J_{cj}^{zz}$ ,  $J_{cj}^{yy} - J_{cj}^{zz}$ and  $K_c^{xx} - K_c^{zz}$  terms and the number of the nearest neighbors for each shell. The MAE shows a preference of in-plane magnetization for CGS, while CGT and CGST present an out-of-plane easy axis. This is consistent with experimental reports for CGT [2]. All compounds are semiconducting, with a band gap well below 1 eV.

Next, we calculated the exchange parameters for these compounds and, in Table III, we present the most relevant values up to the fifth-nearest neighbors. The interactions are given in the local frame of reference shown in Fig. 2(a), while the corresponding pairs of atoms ij are labeled as depicted in Fig. 1(b). The negative value of  $J_{c1}^{H}$  indicates that the interaction between first neighbors is FM, which agrees well with experimental results for CGT [1] and other theoretical results [16,30]. The uniaxial on-site anisotropy energy,  $K_c^{xx} - K_c^{zz}$ , is approximately  $\sim$  15 times larger in CGT than in CGS and 3 times larger than in CGST. This highlights the effect of SOC interaction in heavy atoms such as Te compared to the lighter atoms like Se. On-site anisotropy values for these materials show a preference of in-plane magnetization. However, the two-site exchange anisotropy  $J_{cj}^{xx} - J_{cj}^{zz}$  and  $J_{cj}^{yy} - J_{cj}^{zz}$  also contributes to the total MAE, especially at first neighbors (i = 1). This can be attributed to the exponential decay of the exchange interactions due to the semiconducting behavior of the materials. We find that it is this type of magnetic

TABLE III. Exchange parameters for CGS, CGT, and CGST in the local reference frame given by Fig. 2(a). All quantities are given in meV units. Atomic labels correspond to those defined in Fig. 1(b). Due to  $C_3$  symmetry,  $K_c^{xx} = K_c^{yy}$ . The values for  $S_{c2}^{\alpha}$  are omitted in the table since their magnitudes are below 0.01 meV, with the exception of  $S_{c2}^x$  in CGST, where it is 0.04 meV. Isotropic exchange couplings beyond the fifth-nearest neighbors are below 0.05 meV. The number of first-, second-, third-, fourth-, and fifth-nearest neighbors with respect to the *c* atom are 3, 6, 3, 6, and 6, respectively.

	CGS	CGT	CGST
$\overline{K_c^{xx} - K_c^{zz}}$	-0.03	-0.47	-0.15
$J_{c1}^{H}$	-0.86	-7.04	-4.38
$J_{c1}^{xx} - J_{c1}^{zz}$	-0.01	-0.06	0.03
$J_{c1}^{yy} - J_{c1}^{zz}$	0.09	0.72	0.58
$S_{c1}^x$	-0.01	-0.02	-0.04
$D_{c1}^{y}$			0.70
$D_{c1}^{z}$			-0.19
$J^H_{c2}$	0.28	0.19	0.72
$J_{c2}^{xx} - J_{c2}^{zz}$	< 0.01	-0.02	-0.04
$J_{c2}^{yy} - J_{c2}^{zz}$	-0.01	-0.05	-0.04
$D_{c2}^x$	-0.16	-0.19	-0.16
$D_{c2}^{y}$			0.07
$D_{c2}^{z}$	-0.19	-0.42	-0.25
$J^H_{c3}$	0.30	-0.25	0.24
$J_{c3}^{xx} - J_{c3}^{zz}$	< 0.01	-0.11	-0.12
$J_{c3}^{yy} - J_{c3}^{zz}$	-0.02	0.04	-0.05
$S_{c3}^x$	-0.02	-0.14	-0.10
$D_{c3}^{y}$			-0.02
$D_{c3}^{z}$			-0.28
$J^H_{c4}$	0.21	0.19	0.24
$J_{c5}^H$	0.34	0.37	0.39

anisotropy that leads to out-of-plane magnetization in CGT and CGST.

The only off-diagonal component of the symmetric exchange tensor that doesn't vanish by symmetry is  $S_{c1}^x$ , but its value is very small compared to the isotropic interactions, with no relevant differences among the three structures. Conversely, DM vectors at first-nearest neighbors appear only for the Janus CGST compound due to the breaking of inversion symmetry. Of particular interest is the DM vector component  $D_{c1}^y$  for CGST, with a value that is approximately ~16% of the isotropic exchange  $J_{c1}^H$  and agrees well with VASP supercell calculations [16]. Remarkably, this component could induce the formation of radial vortex magnetic patterns in CGST, as shown in Fig. 2(c).

#### A. Biaxial strain

After describing the magnetic interactions in the ground state, we focus on different effects that could be applied experimentally to modify these interactions, eventually triggering a controllable change in the magnetic response of those systems. We first consider the effect of homogeneous biaxial strain. This is simulated by relaxing the structures for different



FIG. 3. Isotropic exchange constants up to the fourth-nearest neighbors as a function of the biaxial strain for (a) CGS, (b) CGT, and (c) CGST. The labels cj (j = 1, 2, 3, and 4) refer to pairs of sites as depicted in Fig. 1(b).

fixed in-plane lattice constants. With respect to the groundstate geometry, we look for strains in the range of  $\pm 4\%$ . In this range, the easy axis for the three materials remains unchanged, and the magnetic moments vary by approximately 2%, ensuring that the ground state multiplet is not affected by the strain.

Biaxial strain does not affect the symmetry of the system. As a consequence, we find that the most relevant changes happen in the isotropic exchange constants. These are plotted in Fig. 3 as a function of strain. The first-neighbor interactions are the most sensitive on the strain because of the Goodenough-Kanamori-Anderson (GKA) mechanism [50–52]. This phenomenon is related to the angle formed by consecutive Cr-X-Cr bonds (X =Se or Te) that increases with strain. At short Cr-Cr distances, the interaction between

TABLE IV. Angle subtended for the bonds Cr-X-Cr (X=Te and Se) in the three materials for some values of the biaxial strain. For CGST, the bond angles are shown for Cr-Te-Cr and Cr-Se-Cr.

Biaxial strain	CGS	CGT	CGST
-3%	87.61°	88.63°	84.33°/91.52°
0%	89.94°	90.97°	86.98°/94.05°
+3%	92.11°	93.12°	87.72°/94.70°

Cr atoms is antiferromagnetic, driven by the Pauli exclusion principle and the presence of half-filled  $t_{2g}$  orbitals in the Cr<sup>3+</sup> atoms. However, when the Cr-Cr distance increases and the angle bond is ~ 90°, a FM superexchange interaction mediated by the *p* orbitals of Te or Se takes over [28,53,54]. This implies that when the bond angle passes the critical value of ~ 90°, an AFM-to-FM transition occurs. This is due to the competition between the AFM direct exchange and the FM superexchange interactions, with the decay of the AFM direct exchange with Cr-Cr distance being more pronounced than that of the FM superexchange. The same mechanism is found in other magnetic systems such as 1*T*-CrTe<sub>2</sub> [55], CrSiTe<sub>3</sub> [56], or other Janus magnets like Cr<sub>2</sub>(*X*, *Y*)Te<sub>6</sub> (*X*, *Y* = Ge, Si, Sn) [57].

The isotropic Heisenberg interactions for CGS are shown in Fig. 3(a) as a function of the lattice constant. For this compound, a compression of only ~ 0.35% changes the sign of  $J_{1c}^{H}$ to favor AFM interaction. Moreover, a larger compression of the order of ~ 3% results in  $J_{c1}^{H}$  ~ 8 meV, which is about four times larger than the second strongest isotropic coupling. This implies that CGS is a 2D magnet, where the ground state can be switched from FM to AFM experimentally through feasible biaxial compression.

As is clear from Figs. 3(b) and 3(c), for the other two compounds larger compressions, ~2.7% for CGT and ~1.6% for CGST, are needed to have a sign change in  $J_{c1}^{H}$ . In case of a few strain values, in Table IV we listed the angles between the Cr-Se/Te and Se/Te-Cr bonds connecting nearest-neighbor Cr atoms. Regarding  $J_{c1}^{H}$  for these strain values, we find a good agreement with the GKA mechanism. Note that for lattice constants below the FM-AFM transition for  $J_{c1}^{H}$ , all isotropic constants up to the fourth-nearest neighbors become positive (AFM coupling), indicating the loss of FM collinear magnetic order.

Beyond the effect on the isotropic exchange constants, we also noticed significant impacts of biaxial strain on the DM vectors, in particular, for  $\mathbf{D}_{c2}$ . In the considered range of strain,  $D_{c2}^x$  varies between about -0.1 and -0.2 meV in CGT, while  $D_{c2}^z$  varies between -0.1 and -0.2 meV in CGS and between -0.3 and -0.4 meV in CGT.

#### **B.** Uniaxial strain

Next we investigate the effect of uniaxial strain on the exchange interactions. We simulate a strain in the range of  $\pm 5\%$  applied only over the direction of  $\vec{a}_1$ , as defined in Fig. 1(a). This strain breaks  $C_3$  symmetry, therefore, the *c*1 and *c*1' bonds are no longer equivalent. This implies the lifting of degeneracy between the isotropic exchange parameters  $J_{c1}^H$  and  $J_{c1'}^H$ , which can have different characteristics depending

TABLE V. Difference  $\Delta \alpha$  between the angles subtended by the Cr-X-Cr bonds along the *c*1 and *c*1' nearest neighbors, when  $\pm 3\%$  uniaxial strain is applied over the  $\vec{a}_1$  direction. For CGST,  $\Delta \alpha$  is presented for the Cr-Te-Cr/Cr-Se-Cr bonds.

Uniaxial strain	CGS	CGT	CGST
-3%	2.30°	$2.42^{\circ}$	2.18°/2.48°
+3%	-2.33°	-2.45°	-2.22°/-2.31°

on the distance between Cr atoms and, consequently, on the Cr-X-Cr (X = Te, Se) bond angles. The difference between the angles subtended by the c1 and c1' types of Cr-X-Cr bonds,  $\Delta \alpha$ , is shown in Table V. The values of  $\Delta \alpha$  are of the order of the differences between the angles shown in Table IV, indicating that the effect of the GKA mechanism might be very different for the c1 and c1' bonds.

In Fig. 4, the nonequivalent isotropic couplings for the nearest neighbors are shown as a function of the uniaxial strain. All the  $J_{c1'}^H$  curves present similar slopes, considerably larger than those of  $J_{c1}^H$ . This is a consequence of the c1' bond located in the direction of the strain, and therefore being more sensitive to its magnitude. Due to these different slopes, we find regions of compressive strain where  $J_{c1'}^H$  changes signs to AFM coupling but  $J_{c1}^H$  remains FM. Most likely, these strain values would favor a magnetic structure of FM coupled rows of Cr atoms in a direction perpendicular to the strain, with AFM coupling between rows. The needed compression to obtain this transition is of only ~0.5% for CGS, while a larger value of ~2.5% (~4%) is needed for CGST (CGT).

Focusing on the impact of uniaxial strain on the DM vectors, we can note that most bonds retain their symmetries under uniaxial strain. The most obvious exception is  $c^2$ , since upon uniaxial strain the distance from site 1/ to c will differ



FIG. 4. Nearest-neighbor isotropic exchange couplings as a function of uniaxial strain over  $\vec{a}_1$  for CGS (orange), CGT (purple), and CGTS (green). The interactions for the pairs split by the strain, labeled *c*1 and *c*1', show remarkably different dependence on the strain. The labels cj (j = 1 and 1') refer to nearest-neighbor pairs of sites as depicted in Fig. 1(b).

from that between sites 1' to 2' and the  $C_{2y}$  symmetry in the local frame of reference (see Fig. 2(a) and Table I) gets broken. This does not happen for the c2 bond, however. Therefore,  $D_{c2'}^{y}$  is induced by uniaxial strain. The magnitude of  $D_{c2'}^{y}$ induced by  $\pm 5\%$  uniaxial strain is  $\mp 0.1$  meV ( $\mp 0.05$  meV), that is, ~50% (16%) of  $J_{c2'}^{H}$  for CGT (CGS). Recall that  $C_{2y}$ symmetry was already broken in CGST and  $D_{c2'}^{y}$  is different from zero even when uniaxial strain is not applied. No other relevant changes are observed for the relativistic exchange parameters, apart from those observed for biaxial strain.

From an experimental point of view, compression through strain is difficult to achieve without avoiding buckling effects. However, compression values on the order of 0.2% can be obtained [58] and are close to the values needed to achieve the transition in CGS. Furthermore, to increase the compression, more sophisticated techniques may be used, such as sulfur or oxygen substitution [59–61], the introduction of vacancies [62], or other methods [63].

### C. Gate voltage

We simulate the effect of gate voltage by including in our simulations an electric field  $\mathcal{E}_z$  perpendicular to the monolayer. The range of electric field values we are using can be experimentally achieved with an STM tip [64]. The main effect of such an electric field is breaking the inversion symmetry of the CGS and CGT structures which induces DM interactions in the system [27,65-68], while the isotropic and other exchange parameters are hardly affected by the gate voltage. We find that  $\mathcal{E}_z$  produces a spontaneous linear induction in  $D_{c1}^{y}$  for CGS and CGT which was zero in the absence of the field. Inversion symmetry is already broken for CGST without electric field, however, a change in the magnitude of the DM vectors proportional to the electric field is also observed. Figure 5(a) indicates that in CGS and CGT the chirality of the first-nearest-neighbor DM vectors, each parallel to the local y axis, is controlled by the sign of the electric field. In the case of CGST, we find that  $D_{c1}^{y} > 0$ , so the chirality of the DM vectors is the one shown in the right picture in Fig. 5(a) in the investigated range of the electric field. Figure 5(b) demonstrates the linear response in  $D_{c1}^{y}$  to the applied electric field for the three materials. A linear fit, performed by standard methods [69] to the expression  $D_{c1}^{y}(\mathcal{E}_{z}) = \mathcal{P}_{X} \cdot \mathcal{E}_{z}$  shows that  $\mathcal{P}_{CGS} = -0.11 \,\mathrm{e} \cdot \mathrm{\AA}$ ,  $\mathcal{P}_{\text{CGT}} = -0.36 \,\mathrm{e} \cdot \text{\AA}$  and  $\mathcal{P}_{\text{CGST}} = -0.10 \,\mathrm{e} \cdot \text{\AA}$ . The induced  $D_{c1}^{y}$  component can achieve 3% and 1.6% of  $J_{c1}^{H}$  with  $\mathcal{E}_{z} = 0.3$ V/Å for CGS and CGT, respectively. For CGST, the linear response is small compared to the value in the absence of the field, and this induces a total magnitude change of 6% of  $D_{c1}^{y}(\mathcal{E}_{z}=0)$ . Remarkably, in this compound  $D_{c1}^{y}$  is between 15–16% of  $J_{c1}^H$  for  $\mathcal{E}_z \in [-0.3, 0.3]$  V/Å.

The electrically induced DM interaction  $D_{c1}^y$  is further enhanced by biaxial strain. We found that a strain of 3% increases the magnitude of  $D_{c1}^y$  by ~60% when  $\mathcal{E}_z = 0.3 \text{ V/Å}$ is applied for CGS and CGT. Moreover, a biaxial compression of 3% reduces the magnitude of  $D_{c1}^y$  by 30% and 46% for CGS and CGT, respectively.

Lastly, we comment on the potential ferroelectric properties that can be found in Janus magnets [17,70–72] like CGST. We find that the total electric dipole moment of CGS,



FIG. 5. (a) Schematic drawing of the angular vortex pattern of the nearest-neighbor DM vectors depending on the sign of the applied electric field normal to a monolayer of CGS and CGT. (b)  $D_{c1}^y$ as a function of the electric field for CGS (orange), CGT (purple), and CGST (green). CGS and CGT curves cross the origin because of the inversion symmetry of the system when  $\mathcal{E}_z = 0$ , unlike in the case of CGST.

CGT, and CGST per unit cell follows the linear response relationship

$$\mathcal{P}_T = \mathcal{P}_0 + \alpha \, \mathcal{E}_z,\tag{4}$$

where  $\alpha$  is the material's polarizability, see Fig. 6. We find that the *z*-axis charge asymmetry in CGST produces a nonzero  $\mathcal{P}_0 = 0.16 \text{ e}$  Å. The obtained  $\mathcal{P}_0$  in this Janus magnet is about ten times larger than those obtained through bilayer stacking engineering [73,74], but several times smaller than those for perovskite-based compounds [75]. The simultaneous presence



FIG. 6. Total electric dipole moment  $\mathcal{P}_T$  measured in  $e \cdot \text{Å}$  along the *z* axis for the three different materials. In absence of electric field, CGST has a total dipole equal to 0.15  $e \cdot \text{Å}$  that can be switched with electric fields below -0.1 V/Å.

of the electric dipole moment and magnetization in the same phase confirms the multiferroic behavior of CGST. However, since only a few Janus materials have been experimentally synthesized, no experimental measurements have yet been performed to confirm their multiferroic behavior. Furthermore, a negative electric field larger in magnitude than  $\mathcal{E}_z =$ 0.1 V/Å reverses the sign of  $\mathcal{P}_T$ , as shown in Fig. 6. For CGS and CGT,  $\mathcal{P}_0$  is zero. We find that the polarizability is similar for the three compounds. The proportionality of  $D_{c1}^y$ with the electric field seen in Fig. 5(b) is consistent with the linear response  $\mathcal{P}_T \propto \mathcal{E}_z$  [66,76].

#### V. SUMMARY AND CONCLUSIONS

We presented a computational study of the impact of symmetry breaking induced by different experimental approaches on the magnetic properties of 2D vdW materials. We calculated the tensorial exchange parameters for CrGeSe<sub>3</sub>, CrGeT<sub>3</sub>, and Cr<sub>2</sub>Ge<sub>2</sub>(Se, Te)<sub>3</sub> using the program package GROGU [24] based on the relativistic version [46] of the LKAG formalism [45]. CGST exhibits ferroelectric properties and is a candidate to support magnetic textures due to DM interaction arising from the lack of inversion symmetry [16]. Biaxial strain can trigger a FM/AFM transition among first neighbors with experimentally feasible compressions of approximately 0.4, 2, and 3% for CGS, CGST, and CGT, respectively. Uniaxial strain produces a difference between isotropic couplings for the c1 and c1/ types of nearest neighbor bonds, and reasonable values of compression ( $\sim 0.5\%$ , 2.5% and 4% for CGS, CGST and CGT, respectively) results to isotropic couplings  $J_{c1}^{H} > 0$  and  $J_{c1}^{H} < 0$ . These couplings then promote a rowwise AFM texture of the Cr spins observed in other monolayer systems [77,78]. Uniaxial strain breaks  $C_{2y}$  symmetry along the next nearest neighbor c2' bond and induces  $D_{c2'}^{y}$  component that can achieve 50%/16% of  $J_{c2'}^{H}$  for CGT/CGS. A gate voltage perpendicular to the monolayer breaks inversion symmetry for CGS and CGT, and  $D_{c1}^{y}$  emerges in linear response to the electric field, forming a vortex pattern at the first nearest neighbors whose chirality is tuned by the sign of the electric field. Our computational results clearly demonstrate that relevant exchange parameters for the 2D vdW ferromagnets CGS, CGT, and CGST can be tuned by feasible external control to achieve magnetic phase transitions.

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### APPENDIX: SYMMETRY CONSIDERATIONS

Here we summarize the selection rules that occur over the exchange constants of the Heisenberg Hamiltonian in Eq. (2) due to the invariance of the structure under certain symmetry operations. These rules are the extension of Moriya's rules for the DM vectors [79]. The symmetry operations are performed with the origin at the center C of the bond between the atoms A and B, as previously shown in Fig. 2(a). The rules are the following:

(1) Inversion symmetry ( $\Pi$ ) respect to C:

$$D_{AB}^x = D_{AB}^y = D_{AB}^z = 0$$

- (2) *yz*-plane symmetry ( $\sigma_{yz}$ ) passing through C:
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 $S_{AB}^{y} = S_{AB}^{z} = D_{AB}^{x} = 0.$ (3) *zx*-plane symmetry ( $\sigma_{zx}$ ) passing through C:  $S_{AB}^{x} = S_{AB}^{z} = D_{AB}^{x} = D_{AB}^{z} = 0.$ (4) *xy*-plane symmetry ( $\sigma_{xy}$ ) passing through C:  $S_{AB}^{x} = S_{AB}^{y} = D_{AB}^{x} = D_{AB}^{y} = 0.$ (5) *C*<sub>2</sub> symmetry along *z* axis (*C*<sub>2*z*</sub>) passing through C:  $S_{AB}^{x} = S_{AB}^{y} = D_{AB}^{z} = 0.$ (6) *C*<sub>2</sub> symmetry along *y* axis (*C*<sub>2*y*</sub>) passing through C:  $S_{AB}^{x} = S_{AB}^{z} = D_{AB}^{y} = 0.$ (7) *C*<sub>2</sub> symmetry along *x* axis (*C*<sub>2*x*</sub>) passing through C:  $S_{AB}^{y} = S_{AB}^{z} = D_{AB}^{y} = 0.$ (8) *C<sub>n</sub>* (*n* > 2) symmetry along *x* axis (*C<sub>nx</sub>*) passing through C:  $S_{AB}^{y} = S_{AB}^{y} = S_{AB}^{z} = D_{AB}^{y} = D_{AB}^{z} = 0.$ 

 $S_{AB}^{x^{2}} = S_{AB}^{y} = S_{AB}^{z} = D_{AB}^{y} = D_{AB}^{z} = 0$  and  $J_{AB}^{xx} = J_{AB}^{yy}$ . The application of an electric field along the *z* axis or Janus monolayers in the *xy* plane breaks the following symmetries:  $\Pi$ ,  $\sigma_{xy}$ ,  $C_{2x}$ , and  $C_{2y}$ , therefore, the constraints following from rules 1, 4, 6, and 7 do not apply.

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