Zeeman effect in centrosymmetric antiferromagnetic semiconductors controlled by electric field

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Centrosymmetric antiferromagnetic semiconductors, although abundant in nature, seem less promising than ferromagnets and ferroelectrics for practical applications in semiconductor spintronics. As a matter of fact, the lack of spontaneous polarization and magnetization hinders the efficient utilization of electronic spin in these materials. Here, we propose a paradigm to harness electronic spin in centrosymmetric antiferromagnets via Zeeman spin splittings of electronic energy levels – termed as spin Zeeman effect – which is controlled by electric field. By symmetry analysis, we identify twenty-one centrosymmetric antiferromagnetic point groups that accommodate such a spin Zeeman effect. We further predict by first-principles that two antiferromagnetic semiconductors, Fe₂TeO₆ and SrFe₂S₂O, are excellent candidates showcasing Zeeman splittings as large as ~55 and ~30 meV, respectively, induced by an electric field of 6 MV/cm. Moreover, the electronic spin magnetization associated to the splitting energy levels can be switched by reversing the electric field.

Our work thus sheds light on the electric-field control of electronic spin in antiferromagnets, which broadens the scope of application of centrosymmetric antiferromagnetic semiconductors.

Introduction. – In semiconductors, the creation of magnetically or electrically controllable spin splittings with relatively large magnitudes is at the heart of designing semiconductor spintronics devices (e.g., spin transistor) [1–5]. The conventional ferromagnetic or ferroelectric semiconductors naturally host such controllable spin splittings because of the existence of a spontaneous magnetization or polarization, thanks to Rashba-Dresselhaus [4–7], or Zeeman [1, 8] effect. In sharp contrast, the centrosymmetric antiferromagnetic semiconductors do not have any polarization and their magnetization is either null or tiny. The lack of polarization and magnetization makes it challenging to generate a sizable and controllable spin splitting, by magnetic or electric field, in centrosymmetric antiferromagnetic semiconductors. Consequently, the centrosymmetric antiferromagnetic semiconductors – in spite of their abundance in nature – seem not promising for practical applications in spintronics [9–14].

Recently, efforts were made to explore the possible spin splittings hosted by all types of magnetic space groups, involving non-magnetic, ferromagnetic, and antiferromagnetic materials (see, e.g., Refs. [14–21]). Several previously-overlooked spin-splitting patterns were discovered [14–21], but without demonstrating the possibility of creating and controlling sizable spin splittings in centrosymmetric antiferromagnets by magnetic or electric field. Interestingly, two works focusing on nonlinear photocurrent in MnBi₂Te₄ [22] and magneto-optic Kerr effect in MnPSe₃ [23] (rather than spin splittings) hint to such a possibility. However, the general conditions and underlying mechanisms to the creation and control of spin splittings by magnetic or electric field in centrosymmetric antiferromagnets remain elusive.

In this Letter, we aim at exploring spin splittings that would be controllable by electric field and hosted by centrosymmetric antiferromagnets. Our basic idea is rooted in the magnetoelectric effect (see e.g., Ref. [24]). As a matter of fact, electric field not only creates polarization $P_\alpha$ but also generates magnetization $M_\beta \propto P_\alpha$ in magnetoelectric antiferromagnets ($\alpha, \beta = x, y, z$) [24]. The occurrence of $M_\beta$ implies an effective internal magnetic field $B_\beta^{\text{eff}} \propto P_\alpha$ in materials, which couples with electronic spin $\sigma_\beta$ (i.e., Pauli matrix $\sigma_\beta$) and yields a Zeeman-like Hamiltonian $\lambda_{\alpha,\beta} P_\alpha \sigma_\beta$ [25].

Further, we check our idea by symmetry analysis and first-principles simulations. We identify twenty-one centrosymmetric antiferromagnetic point groups that accommodate electrically controllable Zeeman spin splittings. More promisingly, we find two centrosymmetric antiferromagnetic semiconductors, Fe₂TeO₆ and SrFe₂S₂O, in which large Zeeman spin splittings of ~55 and ~30 meV can be created by electric field of 6 MV/cm, respectively. The electronic spin magnetization associated to the splitting energy levels is confirmed to be switchable by reversing the electric field.

Couplings between polarization and spin. – Among 122
magnetic point groups (MPGs), there are eleven groups, namely, 11, 2/m1, mmm1, 4/m1, 4/mmm1, 31, 3m1, 6/m1, 6/mmm1, m31, and m3m1, that contain both inversion \( \bar{1} \) and time-reversal \( \bar{\sigma} \) symmetries. As will be shown below, these eleven groups belong to type-II Shubnikov MPGs (denoted by \( G' \)) – host a sequence of subgroups (i.e., type-III Shubnikov MPGs) that allow the couplings between polarization and spin. Here, \( G' \) can be uniformly written as \( G' = G \cup \bar{G} \), where \( G = G_0 \cup \bar{G}_0 \) is the crystallographic point group and \( G_0 \) is the subgroup of \( G \) containing only proper rotations \( 27, 29 \). We aim at finding the minimal couplings involving electric polarization and spin with respect to \( G' \) group. To this end, we examine the transformation behaviors of electric polarization \( P_\alpha \) and spin angular momentum \( S_\beta \) under \( \bar{1} \) and \( \bar{\sigma} \), where \( \alpha, \beta = x, y, z \) denote the Cartesian components (see Fig. 4). Normally, the coupling \( P_\alpha S_\beta \) (if existing) implies the electronic spin splittings induced by polarization \( P_\alpha \), recalling that spin splitting is characterized by \( \sigma_\beta \) where \( S_\beta = \frac{1}{2} \sigma_\beta \). Figures 1(a)–1(f) indicate the following transformation rules, namely, \( \bar{1} : P_\alpha \rightarrow -P_\alpha, S_\beta \rightarrow S_\beta, \sigma_\beta \rightarrow -\sigma_\beta \) and \( \bar{\sigma} : P_\alpha \rightarrow P_\alpha, S_\beta \rightarrow -S_\beta, \sigma_\beta \rightarrow -\sigma_\beta \). Hence, the bilinear coupling between polarization and spin does not exist in the presence of either inversion or time-reversal symmetry, because \( \bar{1} : P_\alpha \sigma_\beta \rightarrow -P_\alpha \sigma_\beta; \bar{\sigma} : P_\alpha \sigma_\beta \rightarrow -P_\alpha \sigma_\beta \).

We move on to explore whether the trilinear coupling \( X \sigma_\alpha \sigma_\beta \) does exist or not with respect to \( G' \). First, to fulfill the inversion and time-reversal symmetries, \( X \) should be a quantity such that \( \bar{1} : X \rightarrow -X \) and \( \bar{\sigma} : X \rightarrow -X \). Figures 1(g)–1(i) showcase such a possible \( X \) extracted from an antiferromagnetic structure. For demonstrating purpose, we simply assume that the \( X \) quantity, namely, magnetic order parameter, is formed by two atoms labelled by \( X' \) and \( X'' \), where \( X' \) and \( X'' \) are of the same atomic species, but carry magnetic moments along opposite directions [see Fig. 1(g)]. Under inversion \( \bar{1} \), \( X' \) and \( X'' \) atoms swap their positions, while their carried magnetic moments remain unchanged [see Fig. 1(h)]. Under time-reversal \( \bar{\sigma} \), \( X' \) and \( X'' \) atoms remain in place with the magnetic moments being flipped [see Fig. 1(i)]. This leads to \( \bar{1} : X \rightarrow -X \) and \( \bar{\sigma} : X \rightarrow -X \). Therefore, \( X \sigma_\alpha \sigma_\beta \) is compatible with inversion and time-reversal symmetries. Next, \( X \sigma_\alpha \sigma_\beta \) should be allowed by the proper rotation operations in \( G_0 \). The \( \Pi' \) is the simplest case to tackle, because its corresponding \( G_0 \) group only contains identity symmetry. Consequently, nine different couplings \( X \sigma_\alpha \sigma_\beta \) with \( \alpha, \beta = x, y, z \) are permitted by symmetry operations of \( \Pi' \) group. Unfortunately, the situation for the remaining ten type-II Shubnikov MPGs is quite complicated, since \( G_0 \) group contains more symmetry operations than identity, leading to additional symmetry constraint to \( X \sigma_\alpha \sigma_\beta \). For instance, \( z \), rotation of \( \pi \) along \( z \) direction, transforms \( P_\alpha \), \( P_\beta \), and \( \sigma_\zeta \) as \( \bar{1} : P_\alpha \rightarrow P_\alpha, P_\beta \rightarrow -P_\beta, \sigma_\zeta \rightarrow -\sigma_\zeta \). As a result, \( \bar{1} \) transforms \( X \sigma_\alpha \sigma_\beta \) and \( X \sigma_\alpha \sigma_\beta \) via \( \bar{1} : X \rightarrow -X \) and \( \bar{1} : X \rightarrow -X \). Therefore, \( X \sigma_\alpha \sigma_\beta \) is compatible with inversion and time-reversal symmetries. Now we demonstrate how to search for real materials hosting \( X \sigma_\alpha \sigma_\beta \) coupling. First of all, note that the existence of order parameter \( X \) breaks inversion \( \bar{1} \), time-reversal \( \bar{\sigma} \), and/or some other symmetry operations of \( G' \) group. Such symmetry breaking lowers the symmetry of the system from \( G' \) group to its subgroup \( g' \) which contains the operations that are not broken by \( X \). In such sense, \( X \) is invariant under all the symmetry operations of \( g' \). With respect to \( g' \), the effective Hamiltonian term \( \lambda_{\alpha,\beta} X \sigma_\alpha \sigma_\beta \) can be re-written as \( \lambda_{\alpha,\beta} X \sigma_\alpha \sigma_\beta \), noting that the quantity \( X \) is absorbed by
the coefficient $\lambda'_{\alpha,\beta}$. Therefore, to find a real material hosting $X$ order parameter and $\lambda'_{\alpha,\beta}P_\alpha\sigma_\beta$ coupling, effort should be made to search for materials with magnetic point group $g'$. Following this logic, we conduct symmetry analysis for the eleven aforementioned type-II Shubnikov MPGs, in order to extract the possible $g'$ groups from $G'$ (see Section I of the SM for the derivations). In particular, we find twenty-one type-III Shubnikov MPGs that accommodate the $\lambda_{\alpha,\beta}P_\alpha\sigma_\beta$ couplings, as summarized in Table I (see Section I.12 and Table S13 of the SM for more details). Interestingly, our derived Zeeman coupling coefficients (Table I) are similar to the tabulated magnetoelastic tensors [57]. In such sense, our proposed twenty-one MPGs also host the magnetoelectric effect, in agreement with our aforementioned analysis (see Introduction). These MPGs do not have inversion or time-reversal $\theta$, but rather exhibit parity-time symmetry ($i\theta$). In essence, these twenty-one type-III Shubnikov MPGs are centrosymmetric in the four-dimensional spacetime, since the $i\theta$ symmetry operation transforms the spatial-temporal coordinate $(x,y,z,t)$ to $(-x,-y,-z,-t)$. Hence, none of these twenty-one MPGs host spontaneous ferromagnetism or electric polarization. According to the $\lambda_{\alpha,\beta}P_\alpha\sigma_\beta$ coupling, the $i\theta$ symmetry operation is broken in the presence of polarization, yielding Zeeman-type spin splittings. This coincides with the previous symmetry analysis which indicates that the breakdown of parity-time symmetry can generate spin splittings (see, e.g., Refs. [14][16][22][23][58][60]).

Taking $m'm'm'$ as an example, Table I indicates the $\lambda_{x,x}$, $\lambda_{y,y}$, and $\lambda_{z,z}$ couplings, yielding the effective Hamiltonian $H(m'm'm') = \lambda_{x,x}P_x\sigma_x + \lambda_{y,y}P_y\sigma_y + \lambda_{z,z}P_z\sigma_z = \kappa_{x,x}E_xE_x\sigma_x + \kappa_{y,y}E_yE_y\sigma_y + \kappa_{z,z}E_zE_z\sigma_z$, where $E_\alpha$ is the electric field along the $\alpha$ direction [61]. Similarly, the effective Hamiltonians for $4'm'm'm'$ and $3'm'$ are given by $H(4'm'm'm') = \kappa_{x,x}(E_xE_x + E_yE_y) + \kappa_{z,z}E_zE_z\sigma_z$ and $H(3'm') = \kappa_{z,z}(E_xE_x + E_yE_y) + \kappa_{z,z}E_zE_z\sigma_z$, respectively. Note that the splittings predicted by $H(3'm')$ were claimed to be critical for the nonlinear photocurrent effect in topological material MnBi$_2$Te$_4$ [22].

Creating and controlling Zeeman splittings in SrFe$_2$S$_2$O and Fe$_2$TeO$_6$. – Based on Table I we search from the MAGNADATA database [51] for antiferromagnets with Zeeman splittings that can be created and controlled by electric field. Promisingly, we find two antiferromagnetic semiconductors, SrFe$_2$S$_2$O and Fe$_2$TeO$_6$ (see Figs. 2), whose Néel temperatures are both higher than 200 K [62][63]. The corresponding MPGs for SrFe$_2$S$_2$O and Fe$_2$TeO$_6$ are $m'm'm'$ [62] and $4'm'm'm'$ [64], respectively. Employing their ground state magnetic structures [sketched in Figs. 2(b) and 2(d)] and considering spin-orbit interaction, we use first-principles to compute the band structures of SrFe$_2$S$_2$O and Fe$_2$TeO$_6$ without polarization or with polarization created by electric field of 6 MV/cm (see Section III.I of the SM). As shown in Fig. S2 of the SM, the valence band maximum (VBM) of SrFe$_2$S$_2$O are located at the Γ point, and the corresponding spin levels are (i) degenerate for non-polarized, (ii) nearly degenerate for $E_x$-polarized, (iii) slightly split for $E_y$-polarized, and (iv) obviously split for $E_z$-polarized SrFe$_2$S$_2$O material. As for Fe$_2$TeO$_6$, the conduction band minimum (CBM) is at the Γ point, and $E_x$ apparently splits the spin levels at the CBM (see Fig. S5 of the SM) [65]. Our numerical simulations further indicate that the magnitudes of Zeeman spin splittings are in perfect linear relationship with $E_\alpha$ (see Fig. 3). Strikingly, $E_y = 6$ MV/cm and $E_z = 6$ MV/cm generate Zeeman spin splittings of ∼30 meV and ∼55 meV [66], respectively, for the VBM of SrFe$_2$S$_2$O and CBM of Fe$_2$TeO$_6$.

On the other hand, we notice that the spin splittings induced by $E_x$, $E_y$, and $E_z$ in SrFe$_2$S$_2$O exhibit highly distinct characteristics (see Fig. 4). For example, electric field $E_x$ of 6 MV/cm causes nearly null Zeeman spin splitting, implying the smallness of the coupling coefficient $\kappa_{x,x}$ in $H(m'm'm') = \kappa_{x,x}E_xE_x\sigma_x + \kappa_{y,y}E_yE_y\sigma_y + \kappa_{z,z}E_zE_z\sigma_z$. Meanwhile, the Zeeman spin splitting induced by $E_y$ is far larger than that generated by $E_z$ of the same magnitude as $E_y$. For interpretation, we analyze the spin magnetization ($S_x$, $S_y$, $S_z$) associated with the two top most energy sublevels at the Γ point. When polarizing SrFe$_2$S$_2$O by $E_\alpha$ ($\alpha = x,y,z$) electric field, an effective magnetic field $B_{\alpha}^{\text{eff}} \propto E_\alpha$ is created in the material. The $B_{\alpha}^{\text{eff}}$ field couples with $S_\alpha$, causing a Zeeman

| $Y$ | $\lambda_{x,x}$ | $\lambda_{y,y}$ | $\lambda_{z,z}$ | $\lambda_{x,y}$ | $\lambda_{x,z}$ | $\lambda_{y,z}$ |
|---|---|---|---|---|---|---|
| $2'm'$ | $\lambda_{x,x}$ | $\lambda_{y,y}$ | $\lambda_{z,z}$ | $\lambda_{x,z}$ | $\lambda_{y,z}$ |
| $2'/m'$ | $\lambda_{x,x}$ | $\lambda_{y,y}$ | $\lambda_{z,z}$ | $\lambda_{x,z}$ | $\lambda_{y,z}$ |

TABLE I. The couplings that are hosted by twenty-one type-III Shubnikov MPGs. In each $(P_\alpha, \sigma_\beta)$ entry, $\lambda_{\alpha,\beta}$ indicates the coupling $\lambda_{\alpha,\beta}P_\alpha\sigma_\beta$; the “...” implies that the coupling $P_\alpha\sigma_\beta$ is forbidden by symmetry. To better understand this table, we refer the readers to Section I.12 of the SM.
energy proportional to $\pm B_{\text{eff}} S_\alpha$, where the $\pm$ sign characterizes the sublevels whose a spin magnetization component are positive or negative. Polarizing SrFe$_2$S$_2$O by $E_y=6$ MV/cm and $E_z=6$ MV/cm leads to the spin magnetization of $S_y \approx \pm 0.74$ and $S_z \approx \pm 0.08$, respectively. The predominant $S_y$ component implies that the Zeeman spin splitting created by $E_y$ is the most prominent. Our further analysis regarding the orbital-projected spin magnetization for SrFe$_2$S$_2$O can be found in Sections III.2 of the SM.

We now address whether the spin magnetization $S_\alpha$ for SrFe$_2$S$_2$O and Fe$_2$TeO$_6$ are switchable by electric field. To begin with, let us recall our model $H(m'm') = \kappa_{x,x} \mathcal{E}_x \sigma_x + \kappa_{y,y} \mathcal{E}_y \sigma_y + \kappa_{z,z} \mathcal{E}_z \sigma_z$ for SrFe$_2$S$_2$O ($m'm'$ group). In the presence of $\mathcal{E}_y$, the spin levels will be split into two sublevels $E_+ = \kappa_{y,y} \mathcal{E}_y$ (eigenstate being $|+\rangle$) and $E_- = -\kappa_{y,y} \mathcal{E}_y$ (eigenstate being $|-\rangle$), where $\sigma_y |+\rangle = |+\rangle$ and $\sigma_y |-\rangle = -|-\rangle$. The spin magnetization $S_y$ associated with $\kappa_{y,y} \mathcal{E}_y$ and $-\kappa_{y,y} \mathcal{E}_y$ are thus $\frac{1}{2} (|+\rangle \sigma_y |+\rangle + |-\rangle \sigma_y |-\rangle) = \frac{1}{2}$ and $\frac{1}{2} (|-\rangle \sigma_y |+\rangle - |+\rangle \sigma_y |-\rangle) = -\frac{1}{2}$ [68]. When reversing electric field from $\mathcal{E}_y$ to $-\mathcal{E}_y$, the two split sublevels become $E_- = \kappa_{y,y} \mathcal{E}_y$ and $E_+ = -\kappa_{y,y} \mathcal{E}_y$, with the corresponding eigenstates given by $|-\rangle$ and $|+\rangle$; Consequently, the $\kappa_{y,y} \mathcal{E}_y$ and $-\kappa_{y,y} \mathcal{E}_y$ sublevels are linked with the spin magnetization $S_y$ of $\frac{1}{2}$ and $\frac{1}{2}$, respectively. Similarly, our models $H(m'm'm')$ and $H(4/m'm'm') = \kappa_{x,x} (\mathcal{E}_x \sigma_x + \mathcal{E}_y \sigma_y) + \kappa_{z,z} \mathcal{E}_z \sigma_z$ predicts that reversing electric field $\mathcal{E}_y$ will switch the $S_\alpha$ spin magnetization between $\pm \frac{1}{2}$ and $\mp \frac{1}{2}$. To confirm our predictions, we compute the local band structures, along with spin magnetization $S_y$ or $S_z$, for SrFe$_2$S$_2$O and Fe$_2$TeO$_6$ (see Fig. 4), including the spin-orbit interaction [69]. We focus on the local bands around the VBM of SrFe$_2$S$_2$O and the CBM of Fe$_2$TeO$_6$. When reversing the electric field from $\mathcal{E}_y = +6$ MV/cm to $\mathcal{E}_y = -6$ MV/cm (respectively, from $\mathcal{E}_z = +6$ MV/cm to $\mathcal{E}_z = -6$ MV/cm), the $S_y$ for SrFe$_2$S$_2$O (respectively, $S_z$ for Fe$_2$TeO$_6$) is switchable. We further find that the $S_z$ component of SrFe$_2$S$_2$O can also be switched by $\mathcal{E}_z$, although the $S_z$ at the VBM is quite small (see Fig. S4 of the SM) [70]. Those results are in qualitative agreement with our model analysis.

To complete this section, let us comment on the limitation of our models. As mentioned above, our models predict $S_\alpha$ as $\pm \frac{1}{2}$ for SrFe$_2$S$_2$O, under electric field $\mathcal{E}_\alpha$. This is at odds with our first-principles simulations, which give, e.g., $S_y \approx \pm 0.74$ and $S_z \approx \pm 0.08$ for SrFe$_2$S$_2$O polarized by $\mathcal{E}_y=6$ MV/cm and $\mathcal{E}_z=6$ MV/cm, respectively. Note that the value $S_z \approx \pm \frac{1}{2}$ (predicted by our model) can be $\sim 6$ times larger than the first-principles-predicted magnitudes (i.e., $S_z \approx \pm 0.08$ of $\mathcal{E}_z$-polarized SrFe$_2$S$_2$O). Such an inconsistency arises from the fact that our models incorporate only the minimal couplings involving electronic spin, electric field, and a mediated magnetic structure. Other degrees of freedom such as atomic orbitals and electronic wave vectors are neglected. More explicitly, our models consider merely two spin sublevels, while the first-principles calculations consider various degrees of freedom (e.g.,...
FIG. 4. Panels (a) and (b) are local band structures of SrFe$_2$S$_2$O polarized by $E_y = +6$ MV/cm and $E_y = -6$ MV/cm, respectively. Panels (c) and (d) show local band structures of Fe$_2$TeO$_6$ polarized by $E_z = +6$ MV/cm and $E_z = -6$ MV/cm, respectively. The color bar corresponds to $S_y$ or $S_z$. The Fermi level $E_f$ is set as the VBM.

the 3d orbitals of Fe ions), forming a multi-band case. As such, our models could only reveal the electric field induced Zeeman splittings qualitatively (i.e., not quantitatively).

Summary and outlook. – We have shown that electric field can create Zeeman spin splittings in centrosymmetric antiferromagnetic semiconductors belonging to one of the twenty-one MPGs (Table I). By first-principles simulations, we further identify two real materials, Fe$_2$TeO$_6$ and SrFe$_2$S$_2$O, that accommodate Zeeman spin splittings as large as $\sim 55$ and $\sim 30$ meV, respectively, in the presence of electric field of 6 MV/cm. The resulting Zeeman spin splittings are controllable by electric field, and can possibly be detected by some approaches (e.g., optical and transport measurements) that are well-established in spintronics [1, 2, 71].

Acknowledgements. – This research was supported by the National Natural Science Foundation of China under Grants No. T2225013, No. 12274174, No. 12174142, No. 12034009, No. 11874207, the Program for JLU Science and Technology Innovative Research Team, and the Science Challenge Project, No. TZZ2016001. L. B. acknowledges the Vannevar Bush Faculty Fellowship (VBFF) Grant No. N00014-20-1-2834 from the Department of Defense and the MonArk Quantum Foundry supported by the National Science Foundation Q-AMASE-i program under NSF Award No. DMR-1906383. We thank Prof. Y. Wei at Fudan University for the valuable discussion. The calculation was performed in the high-performance computing center of Jilin University.

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Note, however, that the coupling \( \lambda \) does not exist in materials with time-reversal symmetry, as will be shown below.

### Magnetic Point Group Tables

- [Magnetic Point Group Tables](https://www.cryst.ehu.es/cryst/mpoint.html)

### Point Group Tables

- [Point Group Tables](https://www.cryst.ehu.es/rep/point.html)

### Supplementary Material

- [See Supplementary Material which includes symmetry analysis, methods, and some numerical results](https://www.cryst.ehu.es/cgi-bin/crystr/progams/mtensor.pl)

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