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Trompe L'oeil Ferromagnetism

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The characteristics of ferro-(ferri)magnetism with non-zero magnetization include magnetic attraction, magnetic circular dichroism, and magneto-optical Kerr (MOKE), Faraday, and various anomalous Hall-type (Hall, Ettingshausen, Nernst, and thermal Hall) effects. Non-magnetic or antiferromagnetic materials in external electric fields or other environments (called specimen constituents) can share symmetry operational similarity (SOS) with magnetization ($\mathcal{M}$) in relation to broken symmetries. These specimen constituents can be associated with non-zero magnetization and/or show ferromagnetism-like behaviors, so we say that they exhibit Trompe L'oeil Ferromagnetism. Examples include linear magnetoelectric materials such as Cr_2O_3 under electric fields, Faraday effect in chiral materials such as tellurium with current flow, magnetic field induced by the motion of Neel- or Bloch-type ferroelectric walls, and magneto-optical Kerr (MOKE), Faraday effect, and/or anomalous Hall-type effects in certain antiferromagnets such as Cr_2O_3 , MnPS_3 , $\text{Mn}_4(\text{Nb,Ta})_2\text{O}_9$, and $\text{Mn}_3(\text{Sn,Ge,Ga})$. A large number of new specimen constituents having SOS with $\mathcal{M}$ are proposed, and require future experimental verification of their ferromagnetism-like behaviors, and also theoretical understanding of possible microscopic mechanisms.

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INTRODUCTION

Ferro-(ferri)magnets with non-zero magnetic moments has been of great fascination and practical value ever since a piece of lodestone (magnetite, Fe_3O_4) was used as a compass more than 2000 years ago^{1–3}. In fact, the invention and further development of storing information with ferro-(ferri)magnets has been a key component of the microelectronics revolution for the last century³. In addition to their magnetic attraction, ferro-(ferri)magnets can show unique physical phenomena such as various magneto-optical properties (magneto-optical Kerr (MOKE), Faraday, and magnetic circular dichroism) and anomalous Hall-type effects^{4–16}. The anomalous Hall-type effects include anomalous Hall, anomalous Ettingshausen, anomalous Nernst, or anomalous thermal Hall effects^{11–16}. It turns out that these phenomena that traditionally thought to occur only in ferro-(ferri)magnets with non-zero magnetic moments can take place in certain specimens with zero magnetic moment, sometimes in the presence of external electric/strain fields or with time evolution. We call these cases as Trompe L'oeil Ferromagnetism.

It turns out that observable physical phenomena can occur when specimen constituents (i.e., lattice distortions or spin arrangements in external fields or other environments, etc., and also their time evolution) and measuring probes/quantities (i.e., propagating light, electrons or other particles in various polarization states, including light or electrons with spin or orbital angular momentum, bulk polarization or magnetization, etc., and also experimental setups to measure, e.g., Hall-type effects) share symmetry operational similarity (SOS) in relation to broken symmetries¹⁷. This SOS relationship includes when specimen constituents have more, but not less, broken symmetries than measuring probes/quantities do. In other words, in order to have a SOS relationship, specimen constituents “cannot have higher symmetries” than measuring probes/quantities do. The power of the SOS approach lies in providing simple and physically transparent views of otherwise unintuitive phenomena in complex materials. Furthermore, this approach can be leveraged to identify

new materials that exhibit potentially desired properties as well as new phenomena in known materials.

In this paper, we discuss that certain specimen constituents with non-magnetic or antiferromagnetic materials can exhibit SOS with magnetization ($\mathcal{M}$), and what kinds of ferromagnetism-like behaviors these specimen constituents can exhibit. Here, we define $\mathbf{R} = \pi$ (i.e. twofold) rotation operation with the rotation axis perpendicular to the $\mathcal{M}$ direction, $\mathbf{R} = \pi$ (i.e. twofold) rotation operation with the rotation axis along the $\mathcal{M}$ direction, $\mathbf{I}$ = space inversion, $\mathbf{M}$ = mirror operation with the mirror perpendicular to the $\mathcal{M}$ direction, $\mathbf{M}$ = mirror operation with the mirror plane containing the $\mathcal{M}$ direction, $\mathbf{T}$ = time reversal operation. (In the standard crystallographic notations, “ $\mathbf{R}$, $\mathbf{R}$, $\mathbf{I}$, $\mathbf{M}$, $\mathbf{M}$, and $\mathbf{T}$ ” are “ $C_{2\perp}$, $C_{2\parallel}$, $\bar{1}$, $m_{\perp}$, $m_{\parallel}$, $1'$ ”, respectively. Our notations are intuitive to consider one-dimensional (1D) objects such as $\mathcal{M}$.) Evidently, $+\mathcal{M}$ becomes $-\mathcal{M}$ by $\mathbf{R}$ symmetry operation, i.e. $\mathcal{M}$ has broken $\mathbf{R}$ symmetry. Similarly, $+\mathcal{M}$ becomes $-\mathcal{M}$ by $\mathbf{M}$ or $\mathbf{T}$ symmetry operation. Note that $+\mathcal{M}$ remains to be $+\mathcal{M}$ under $\mathbf{I}$ symmetry operation. In fact, $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$ is the set of all “independent” broken symmetries of $\mathcal{M}$. Emphasize that since we consider the 1D nature of $\mathcal{M}$, translational symmetry is ignored. The complete sets of broken symmetries for four vectors discussed in this paper are listed in Fig. 1. Electric polarization ($\mathbf{P}$) has broken $\{\mathbf{R}, \mathbf{I}, \mathbf{M}\}$, and velocity vector ($\mathbf{k}$, linear momentum or wave vector) has broken $\{\mathbf{R}, \mathbf{I}, \mathbf{M}, \mathbf{T}\}$. A uniform strain gradient ($\mathbf{SG}$) along the strain gradient direction behaves like $\mathbf{P}$ in terms of symmetry operations; in other words, $\mathbf{SG}$ has broken $\{\mathbf{R}, \mathbf{I}, \mathbf{M}\}$. The act of applying an external electric field ($\mathbf{E}_{\text{ext}}$), inducing electric current (induced $\mathbf{J}$), for various transport measurements where quasi-equilibrium processes are involved has broken $\{\mathbf{R}, \mathbf{I}, \mathbf{M}, \mathbf{T}\}$. Note that $+\mathbf{E}_{\text{ext}}$ (induced $+\mathbf{J}$) becomes $-\mathbf{E}_{\text{ext}}$ (induced $-\mathbf{J}$) under any of $\{\mathbf{R}, \mathbf{I}, \mathbf{M}\}$, but under $\mathbf{T}$ symmetry operation, “induced $+\mathbf{J}$ ” may not become “induced $-\mathbf{J}$ ” while $+\mathbf{E}_{\text{ext}}$ becomes $-\mathbf{E}_{\text{ext}}$. This is because $\mathbf{T}$ operation on “the act of applying an external electric field $+\mathbf{E}_{\text{ext}}$ ” results in “the act of applying an external electric field $-\mathbf{E}_{\text{ext}}$ ”, which does not necessarily accompany “induced $-\mathbf{J}$ ”, and the magnitude of induced $\mathbf{J}$ may change when the direction of $\mathbf{E}_{\text{ext}}$ is switched.

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Fig. 1 Broken symmetries of various vectors. k : velocity vector (linear momentum, wave vector), P : electric polarization (SG : uniform strain gradient vector), M : magnetic magnetization, and E_{ext} : the act of applying the external electric field that tends to induce electric current (Induced J).

Therefore, T operation on $+E_{ext}$ (induced $+J$) results in $-E_{ext}$ (induced $-J$), in general^{18,19}. Also note that periodic crystallographic or magnetic lattices can have two-, three-, four-, and sixfold rotational symmetries (C_2 , C_3 , C_4 , and C_6 symmetries in the standard crystallographic notations, respectively), but 1D measuring probes/quantities such as M , P , and k do not have C_3 , C_4 , and C_6 symmetries around the rotation axis perpendicular to M , P , or k . Thus, specimen constituents having SOS with M , P , and k should not have any of C_3 , C_4 , and C_6 symmetries around the rotation axis perpendicular to M , P , or k . This consideration becomes important when we discuss antiferromagnetic states having SOS with M .

RESULTS AND DISCUSSION

Linear magnetoelectricity

Linear magneto-electrics are antiferromagnetic materials that do not exhibit SOS with P with broken $\{R, I, M\}$ in zero applied magnetic field (H), but do show SOS with P in non-zero H . These linear magneto-electrics exhibit reciprocal magneto-electric effects, i.e., they do not exhibit SOS with M with broken $\{R, M, T\}$ in zero applied electric field (E), but do show SOS with M in non-zero E , and indeed exhibit non-zero M in non-zero E . The left-hand-side specimen constituents in Fig. 2a–c in zero E show only a part of broken $\{R, M, T\}$, but they in non-zero E have now broken all of $\{R, M, T\}$, so show SOS with M , which is consistent with, for example, the linear magnetoelectric effects in Cr_2O_3 ; diagonal linear magnetoelectric effect, corresponding to Fig. 2a, and off-diagonal linear magnetoelectric effect, corresponding to Fig. 2b, before and after spin flop transition, respectively²⁰. The specimen constituents in Fig. 2c are for magnetic monopoles (1st and 2nd), magnetic toroidal moment (3rd), and magnetic quadrupole (4th and 5th). None of the cases in Fig. 2c in zero E has SOS with M , but all of them do have SOS with M in non-zero E ; thus, all can exhibit linear magnetoelectricity. Note that the 1st and 2nd cases in Fig. 2c correspond to N-type magnetic monopole, and S-type magnetic monopole in the presence of the same E will result in $-M$. Similarly, the 3rd case in Fig. 2c corresponds to counter-clockwise magnetic toroidal moment, and clockwise magnetic toroidal moment in the presence of the same E will result in $-M$. We can also consider various spin configurations on (buckled) honeycomb lattice as shown in Fig. 3a–c. None of these spin configurations in zero E has SOS with M , but all of them in non-zero E do have SOS with M , which indicates that all can be linear magnetoelectrics. Most of these cases have not been experimentally observed in real compounds, and it will be highly demanding to verify these symmetry-driven predictions in real materials²¹. These antiferromagnetic states in Fig. 3 have been

Fig. 2 Various antiferromagnetic states in electric field (E) having SOS with M . Various linear magnetoelectric (antiferromagnetic) systems, which exhibit M in the presence of an electric field (E). All blue arrows are spins or M , and all red arrows are E (or P). They also exhibit M in the presence of SG (i.e. show flexo-magnetism). Conversely, all of them exhibit P in the presence of a magnetic field. **a**, **b** correspond to Cr_2O_3 before and after spin flop transition, respectively. **c** magnetic monopole for the first two cases, toroidal moment for the 3rd case, and magnetic quadrupole for the last two cases.

Fig. 3 Various antiferromagnetic states in (buckled) honeycomb lattice in E , having SOS with M . Blue + and – represents out-of-plane spins, and blue arrows are in-plane spins. Black open and closed circles are spins displaced in the opposite directions along the out-of-plane direction; in other words, black open and closed circles form a buckled honeycomb lattice. Green dashed-line and solid-line circles are anions (e.g., oxygen or sulfur ions) displaced in the opposite directions along the out-of-plane direction.

reported in various compounds with (buckled) honeycomb lattice such as $(Mn, Fe, Co, Ni)P(S, Se)_3$, $BaNi_2V_2O_8$, $(Ca, Sr)Mn_2Sb_2$, $Na_2Ni_2TeO_6$, and $(Mn, Co)_4(Nb, Ta)_2O_9$, but their linear magnetoelectricity has mostly not been reported yet^{21–28}. For example, the 2nd case in Fig. 3b corresponds to the Ising antiferromagnetic state in, e.g., $Mn_4(Nb, Ta)_2O_9$ with buckled honeycomb lattice, and the 1st case in Fig. 3c represents the in-plane antiferromagnetic order in buckled honeycomb lattice with anions in, e.g., $MnPSe_3$.

One exemplary system relevant to Fig. 2c is hexagonal (h -) R $(Mn, Fe)O_3$ (R = rare earths), which is an improper ferroelectric with the simultaneous presence of Mn/Fe trimerization in the ab plane and ferroelectric polarization along the c axis. A number of different types of magnetic order with in-plane Mn spins have

been identified in h -R(Mn,Fe)O₃. The so-called A1-type magnetic order in h -R(Mn,Fe)O₃ combined with Mn trimerization can induce a net toroidal moment, and the so-called A2-type magnetic order in h -R(Mn,Fe)O₃ combined with Mn trimerization can accompany a net magnetic monopole^{29,30}. The linear magnetoelectric effect associated with these magnetic monopole and toroidal moment can be a topic for the future investigation. Note that reversing the $\mathbf{E}$ direction in Figs. 2a–c and 3a–c (for example, by $\mathbf{R}$ operation) should accompany the reversal of $\mathcal{M}$, which is consistent with the nature of linear magnetoelectricity. $\mathbf{E}$ like $\mathbf{P}$ has broken $\{\mathbf{R}, \mathbf{I}, \mathbf{M}\}$, and similarly a strain gradient vector ($\mathbf{SG}$, a uniform strain gradient along a particular direction) has broken $\{\mathbf{R}, \mathbf{I}, \mathbf{M}\}$. Thus, when $\mathbf{E}$ is replaced by $\mathbf{SG}$, the left-hand-side specimen constituents in Figs. 2a–c and 3a–c have broken $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$, so do have SOS with $\mathcal{M}$, which means that all linear magnetoelectrics can exhibit flexo-magnetism, i.e. the induction of a net magnetic moment by a strain gradient (the converse may not be true.). We emphasize that there exists a sign ambiguity in all linear magneto-electrics discussed above as well as all cases having SOS with 1D objects in this paper. In other words, the symmetry arguments cannot determine the absolute sign of $\mathcal{M}$, e.g., in Figs. 2 and 3. However, all antiferromagnetic spins in, e.g., the specimen constituent of Fig. 1a flip by 180° through, e.g., $\mathbf{T}$, then the sign of $\mathcal{M}$ also changes. Note that while symmetry does not fix the absolute sign, but microscopic mechanism determines it. For example, the sign of spin-orbital coupling can fix the absolute sign of induced $\mathcal{M}$.

Dynamic or quasi-equilibrium mechanisms for Trompe L'oeil Ferromagnetism

It is also interesting to consider how to induce magnetization in non-magnetic materials in non-trivial manners. Note that since any static configuration of $\mathbf{P}$ or structural distortions cannot break $\mathbf{T}$, a time component has to be incorporated into $\mathbf{P}$ configurations or structural distortions to induce a SOS relationship with $\mathcal{M}$. Two structural examples having SOS with $\mathcal{M}$ are shown in Fig. 4a, b. When electric current is applied to a tellurium crystal with a

Fig. 4 Various specimen constituents having SOS with $\mathcal{M}$. **a** Golden springs represent mono-axial chiral crystallographic structures. **b** $t = -1, 0$, and $+1$ show time evolution. Moving three types of ferroelectric walls (Neel, Bloch, and Ising) are depicted. **c** A Hall-effect-type transport measurement set-up with a plate-shape specimen, where quasi-equilibrium processes are involved, has SOS with $\mathcal{M}$. Induced J is an induced charge current and thermal current under applied E_{ext} and ΔT , respectively, and $(+)$, $(-)$ and (h) , (c) denote the induced electric voltage and thermal gradient, respectively.

screw-type (so called mono-axial) chiral lattice, corresponding to the left-hand-side cartoon of Fig. 4a, $\mathcal{M}$ can be induced in a linear fashion³¹. Consistently, Faraday rotation of linearly-polarized THz light propagating along the chiral axis of a tellurium crystal is also observed in the presence of quasi-equilibrium electric current flow along the chiral axis, and this induced Faraday rotation effect is linearly proportional to the electric current³². This effect is supposed to occur even when light propagation and electric current are parallel, but perpendicular to the chiral axis, as displayed in the right-hand-side cartoon of Fig. 4a, which needs to be verified experimentally. Interestingly, the transfer of photo-current in chiral DNA turns out to be highly spin polarized³³. All of these highly non-trivial effects correspond to the SOS relationship in Fig. 4a. Figure 4b represent rotating $\mathbf{P}$ having SOS with $\mathcal{M}$, and can be realized when a Neel- or Bloch-type ferroelectric wall moves in the direction perpendicular to the ferroelectric wall^{34,35}, even though any motion of Ising-type ferroelectric walls will not induce $\mathcal{M}$. These effects can be utilized to flip the magnetization of a ferro-(ferri)magnetic island sitting on the top of a ferroelectric with Neel- or Bloch-type walls, but have not been experimentally realized yet.

Anomalous Hall-type effects, involving quasi-equilibrium processes, can be also understood in terms of SOS with $\mathcal{M}$. The sets of $(E_{ext}, +, -)$, (E_{ext}, h, c) , $(\Delta T, +, -)$, and $(\Delta T, h, c)$ of Fig. 4c correspond to the Hall, Ettingshausen, Nernst, and thermal Hall effects, respectively. In the case of, e.g., Hall effect, switching both E_{ext} and $(+,-)$ simultaneously can be achieved by any of $\{\mathbf{R}, \mathbf{I}\}$ (here, symmetry operations are defined with respect to $\mathcal{M}$ in the following), so when a specimen with broken $\{\mathbf{R}, \mathbf{I}\}$ can exhibit a Hall effect that is non-linear with E_{ext} ¹⁷. Consider, for example, a Hall effect experiment on “a specimen with not-broken $\mathbf{R}$ or $\mathbf{I}$ ” in external $\mathbf{H}$ or with internal $\mathcal{M}$. Then, the not-broken $\mathbf{R}$ or $\mathbf{I}$ operation on the entire experimental set-up including the specimen results in flipping the sign of E_{ext} and $(+, -)$ Hall voltage without changing $\mathbf{H}/\mathcal{M}$ direction, meaning that the flipping the sign of E_{ext} leads to flipping the Hall voltage sign without changing the magnitude, so the leading term of Hall voltage proportional to E_{ext} is allowed, but a $(E_{ext})^2$ term is not permitted in terms of symmetry. By the way, this works only for $\mathbf{R}$ or $\mathbf{I}$ operation. Thus, if both $\mathbf{R}$ or $\mathbf{I}$ are broken, then no symmetry limits the Hall effect linear in E_{ext} , which opens up the possibility of a non-linear effect such as Hall voltage varying like $(E_{ext})^2$ in terms of symmetry. We here consider only specimens with not-broken $\{\mathbf{R}, \mathbf{I}\}$, i.e. the Hall-type effects linear with E_{ext} . Then, one can show readily that the entire experimental set-up with a specimen with no broken symmetry in the left-hand-side cartoon in Fig. 4c have broken $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$, so do have SOS with $\mathcal{M}$. This leads to a highly non-trivial theorem that any specimen with broken $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$, but not-broken $\{\mathbf{R}, \mathbf{I}\}$ (i.e., having SOS with $\mathcal{M}$) can show all of anomalous Hall, anomalous Ettingshausen, anomalous Nernst, and anomalous thermal Hall effects, which are linear with E_{ext} . This important theorem is closely relevant to the presence of anomalous Hall-type effects in antiferromagnets without any significant net moments, but with SOS with $\mathcal{M}$, which we will discuss later.

MOKE and Faraday-type effects

When an object with angular momentum transmits through a specimen constituent, its behavior can depend on the sign of the angular momentum. This is, in general, called circular dichroism, and the angular momentum can be spin angular momentum (in, e.g., circularly-polarized light) or orbital angular momentum (in, e.g., vortex beams)^{17,36–38}. This angular momentum-sign-dependent (circular in the case of circularly-polarized light) dichroism can be linear (frequency conserving) or non-linear (frequency changing)^{4–10}. Two particular examples of linear circular dichroism are the rotation of light polarization in the

Table 1. The properties of MOKE, Faraday-type effect, and anomalous Hall-type effect in various specimens.

	MOKE	Faraday-type effect	Anomalous Hall-type effect
Chiral structures	No	Yes	No
Cr_2O_3 , MnPSe_3 , $\text{Mn}_4(\text{Nb}, \text{Ta})_2\text{O}_9$	Yes	No	No
Ferro-(ferri)magnets	Yes	Yes	Yes
Antiferromagnets having SOS with $\mathcal{M}$ (e.g., $\text{Mn}_3(\text{Sn}, \text{Ge}, \text{Ga})$)	Yes	Yes	Yes

Here, Faraday-type effect includes the standard Faraday effect as well as circular dichroism, and anomalous Hall-type effects include anomalous Hall, anomalous Ettingshausen, anomalous Nernst, and anomalous thermal Hall effects.

transmission of linearly polarized light through a chiral material (called natural optical activity), and the rotation of light polarization in the transmission of linearly-polarized light through a ferro-(ferri)magnet (called Faraday effect), and these effects result from the different refractive indices or speeds of light of left-circularly-polarized and right-circularly-polarized lights. The non-linear circular dichroism is associated quasi-equilibrium states with inelastic (dissipative) processes, and the circular dichroism in ferro-(ferri)magnetic materials or non-magnetic (or antiferromagnetic) materials in magnetic fields is called magnetic circular dichroism. X-ray magnetic circular dichroism (XMCD) is a non-linear effect, is associated with electronic transitions among atomic orbital states, and has been heavily studied to explore element-specific magnetism of ferro-(ferri)magnets. A giant optical rotation at a THz-frequency-range antiferromagnetic resonance can occur in the collinear antiferromagnetic state of chiral Ni_3TeO_6 , which exhibits a significant natural optical activity in visible optical range at temperatures even far above an antiferromagnetic transition temperature^{39,40}. It turns out that the symmetry requirement for all of the above linear or non-linear effects in transmission- or absorption-type experiments in chiral materials, ferro-(ferri)magnets or even certain antiferromagnets is broken $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{I} \otimes \mathbf{T}\}$ ¹⁷. Therefore, in this paper, all of the above effects, including, e.g., natural optical activity in chiral materials, and Faraday effect and XMCD in magnetic materials, will be called “Faraday-type effects” or just “Faraday effects” for the sake of simplicity.

On the other hand, Magneto-optical Kerr effect (MOKE) refers the light-polarization rotation of linearly polarized light when it is “reflected” on, e.g., ferro-(ferri)magnetic surfaces¹⁷. The symmetry requirement for MOKE is broken $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{T}\}$ ¹⁷. Certainly, ferro-(ferri)magnets with $\mathcal{M}$ does have broken $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{T}\}$, so do exhibit MOKE for light propagation along the $\mathcal{M}$ direction. Chiral material without magnetism can exhibit Faraday effect (in fact, natural optical activity), but does not exhibit MOKE since $\mathbf{T}$ is not broken in chiral materials (see Table 1). Interestingly, antiferromagnetic Cr_2O_3 in Fig. 2a in zero H has broken $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{T}\}$, so does show MOKE. However, both $\mathbf{T}$ and $\mathbf{I}$ are broken in Cr_2O_3 , but $\mathbf{I} \otimes \mathbf{T}$ is not broken, so Cr_2O_3 does not exhibit Faraday effect (see Table 1)⁴¹.

Now, we can prove a second theorem that breaking all of $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{I} \otimes \mathbf{T}\}$ and $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{T}\}$ in a specimen constituent, where $\mathbf{T}$ and $\mathbf{I} \otimes \mathbf{T}$ are broken in the same way, is identical with breaking $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$, so the specimen constituent shows SOS with $\mathcal{M}$. We consider the specimen constituents where $\mathbf{T}$ and $\mathbf{I} \otimes \mathbf{T}$ are broken in the same way, so $\mathbf{I} = \mathbf{I} \otimes \mathbf{T} \otimes \mathbf{T} = \mathbf{T} \otimes \mathbf{T}$, so $\mathbf{I}$ is not broken. When all $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{T}, \mathbf{I} \otimes \mathbf{T}\}$ are broken, but $\mathbf{I}$ is not broken, $\mathbf{R} = \mathbf{I} \otimes \mathbf{M}$ is broken. Furthermore, $\mathbf{M} \otimes \mathbf{R} = \mathbf{I} \otimes \mathbf{R} \otimes \mathbf{R} = \mathbf{R}^{-1} = \mathbf{R}$. Thus, $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$ is the set

Fig. 5 MOKE and Faraday effect in various antiferromagnetic states. **a–d** represent the antiferromagnetic state in Cr_2O_3 , Ising A-type antiferromagnetic order in a honeycomb-lattice bilayer, which can be realized in a bilayer of, e.g., CrI_3 or CrBr_3 , Ising antiferromagnetic order in buckled honeycomb lattice in, e.g., $\text{Mn}_4(\text{Nb}, \text{Ta})_2\text{O}_9$, and in-plane antiferromagnetic order in buckled honeycomb lattice with anions in MnPSe_3 , respectively. All of **a–d** exhibit MOKE, but no Faraday effect. **e–g** depict an inversion-symmetric antiferromagnetic order in kagome lattice, Ising A-type antiferromagnetic order in a heterogeneous honeycomb-lattice bilayer with, e.g., one CrI_3 layer and the other CrBr_3 layer, and Ising A-type antiferromagnetic order in a heterogeneous honeycomb-lattice bilayer. **e–g** should exhibit all of MOKE and Faraday effect as well as anomalous Hall-type effects. The relevant directions for MOKE and/or Faraday effect are the left-right direction in **a** and **e** and the out-of-page-plane direction in **b**, **c**, **d**, **f**, and **g**.

of all independent broken symmetries. Therefore, we can conclude this non-trivial theorem: all materials having SOS with $\mathcal{M}$ exhibit both Faraday effect and MOKE, and any inversion-symmetric materials exhibiting both Faraday and MOKE do show SOS with $\mathcal{M}$. The combination of two theorems in this paper leads to the conclusion that any specimens having SOS with $\mathcal{M}$ can exhibit all of Faraday effect, MOKE, and anomalous Hall-type effects.

A number of antiferromagnetic states without any net (significant) magnetic moments showing MOKE and/or Faraday effect are illustrated in Fig. 5. The antiferromagnetic states in Fig. 5a–c have broken $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{T}\}$, but not-broken $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{I} \otimes \mathbf{T}\}$ (here, symmetry operations are defined along the left-right direction in Fig. 5a and the out-of-page-plane direction in Fig. 5b–d), so are supposed to exhibit MOKE, but no Faraday effect. Figure 5a, identical with Fig. 2a when $\mathbf{E}$ is zero, corresponds to the antiferromagnetic state of Cr_2O_3 . The spin configurations in Fig. 5c, d are reported to be realized in $\text{Mn}_4(\text{Nb}, \text{Ta})_2\text{O}_9$ with buckled honeycomb lattice and MnPSe_3 with buckled honeycomb lattice, respectively^{22,23,28}. Figure 5b represents Ising A-type antiferromagnetic order in a honeycomb-lattice bilayer, which can be realized in a bilayer of CrI_3 or CrBr_3 . MOKE in Cr_2O_3 has been observed, but none of other effects have been reported⁴¹. The antiferromagnetic states in Fig. 5e–g have broken $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{T}\}$, and also broken $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{I} \otimes \mathbf{T}\}$ (here, symmetry operations are defined along the left-right direction in Fig. 5e and the out-of-page-plane direction in Fig. 5f, g), so should exhibit both MOKE and Faraday effect. Figure 5e corresponds to an inversion-symmetric antiferromagnetic state in kagome lattice, which is realized in, for example, Mn_3Sn , Fig. 5f can be Ising A-type antiferromagnetic order in a heterogeneous honeycomb-lattice bilayer with, e.g., one

Fig. 6 Various x-ray circular dichroism. Schematics of **a** XMCD, **b** XNCD, and **c** XMxD on specimens with (induced) magnetization and chirality. The 3rd equality in **b** and the equality in **c** result from mirror (**M**) operation on some of the terms.

CrI₃ layer and the other CrBr₃ layer, and Fig. 5g represents Ising A-type antiferromagnetic order in a heterogenous honeycomb-lattice bilayer. MOKE in Mn₃Sn has been observed, but any other effects have not been reported yet^{42–45}. Note that according to the first theorem discussed earlier, all cases in Fig. 5e–g have broken $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{T}\}$ and $\{\mathbf{M}, \mathbf{M} \otimes \mathbf{R}, \mathbf{I} \otimes \mathbf{T}\}$, so do have SOS with $\mathcal{M}$ (along the left-right direction in Fig. 5e and the out-of-page-plane direction in Fig. 5f, g), and all can also exhibit anomalous Hall-type effects.

Faraday-type effect with x-ray illumination can be observed in x-ray absorption near-edge spectra (XANES) with circularly polarized x-ray. By recording the XANES spectra with left-circularly-polarized σ^+ and right-circularly-polarized σ^- x-ray with a magnetic field applied either parallel (H^+) or antiparallel (H^-) to the x-ray wavevector, we could obtain the three relevant types of dichroism:

$$\text{XMCD} \propto \{\mu(\sigma^-, H^+) - \mu(\sigma^+, H^+)\} - \{\mu(\sigma^-, H^-) - \mu(\sigma^+, H^-)\}.$$

$$\text{XNCD} \propto \{\mu(\sigma^-, H^+) - \mu(\sigma^+, H^+)\} + \{\mu(\sigma^-, H^-) - \mu(\sigma^+, H^-)\}.$$

$$\text{XMxD} \propto \{\mu(\sigma^-, H^+) + \mu(\sigma^+, H^+)\} - \{\mu(\sigma^-, H^-) + \mu(\sigma^+, H^-)\}.$$

where $\mu(\sigma, H)$ stands for the absorption measured for the indicated polarization and sign of the magnetic field. XM(N)CD indicate x-ray magnetic (natural) circular dichroism signals and XMxD is x-ray magneto-chiral dichroism. The XNCD signal is independent of the applied magnetic field, whereas XMCD changes its sign when the direction of the applied magnetic field is reversed. XMxD does not require polarized light, and manifests as only changes in absorption for two directions of magnetic field. These three dichroism effects are pictorially depicted in Fig. 6. Here, $\mathcal{M}$ is magnetization induced by applied H , and golden springs represent left- or right-type structural chirality of a specimen. Note that XMCD becomes zero when $\mathcal{M}$ (or an object having SOS with $\mathcal{M}$) vanishes. If Left=Right (i.e. no chirality), then XNCD vanishes. XMxD is non-zero only if both Left $\neq$ Right (i.e., having chirality) and also $\mathcal{M} \neq 0$. It is anticipated that new types of XMCD, XNCD, and XMxD experiments can be performed on some of specimen constituents discussed in this paper. For example, XMCD is expected in antiferromagnetic specimens having SOS with $\mathcal{M}$: examples include Mn₃(Sn, Ge, Ga) (see below for Mn₃(Ge, Ga)) and the specimens in Fig. 5f, g. XMxD is anticipated in conical spin states or helical spin states in H.

Fig. 7 SOS relationship of various antiferromagnetic states with $\mathcal{M}$. **a**, **b**, **f**, and **g** Various antiferromagnetic states on kagome or AB-stacked triangular lattices having SOS with $\mathcal{M}$. **b** The left-hand-side antiferromagnetic state on kagome lattice does not have SOS with $\mathcal{M}$ due to threefold rotational symmetry around the out-of-plane direction; however, in the presence of uni-axial uniform strain (green double arrow), which breaks the threefold rotational symmetry, it shows SOS with $\mathcal{M}$. **c**, **d** The all-in-all-out magnetic states in kagome lattice and pyrochlore lattice, respectively, in the presence of strain (green double arrow) exhibit SOS with $\mathcal{M}$. **e** The two-in-one-out (or one-in-two-out) spin arrangement in a triangle, which can be a part of antiferromagnetic kagome lattice, and three-in-one-out (or one-in-three-out) spin arrangement in a tetrahedron, which can be a part of antiferromagnetic pyrochlore lattice, do have SOS with $\mathcal{P}$ with broken $\{\mathbf{R}, \mathbf{I}, \mathbf{M}\}$. All of these antiferromagnetic states having SOS with $\mathcal{M}$ can exhibit MOKE, Faraday effect, and anomalous Hall-type effect. Green dots are inversion centers, and green dashed lines depict magnetic unit cells.

Antiferromagnetic states with Trompe L'oeil Ferromagnetism

Herein, we discuss further antiferromagnetic states having SOS with $\mathcal{M}$. The 1st spin configuration in kagome lattice in Fig. 7a, identical with that in Fig. 5e, has broken $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$, so has SOS with $\mathcal{M}$. In fact, both cases in Fig. 7a do have broken $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$, so has SOS with $\mathcal{M}$ and can exhibit MOKE, Faraday effect, and also anomalous Hall-type effects. Indeed, MOKE, anomalous Hall effect (AHE), anomalous Nernst effect, and anomalous thermal Hall effect

have been observed in Mn_3Sn where the 1st antiferromagnetic state in Fig. 7a occurs^{44–53}. The microscopic mechanism for these effects has been proposed to be Berry curvature, providing a fictitious magnetic field, in the unique antiferromagnetic state. Note that anomalous Ettingshausen effect can occur in Mn_3Sn , but has not been reported yet as far as the author is aware of. The antiferromagnetic state on kagome lattice in Fig. 7b has inversion symmetry, but also has threefold rotation symmetry around the out-of-page-plane axis, so does not have SOS with $\mathcal{M}$. However, in the presence of uniform strain (green double arrow), which breaks the threefold rotational symmetry, the Fig. 7b state now shows SOS with $\mathcal{M}$. Similarly, the all-in-all-out magnetic states in kagome lattice and pyrochlore lattice in Fig. 7c, d, respectively, in the presence of uni-axial uniform strain (green double arrow) exhibit SOS with $\mathcal{M}$. In the absence of green arrows, all of Fig. 7b–d have broken $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$, but Fig. 7b, c have threefold rotational symmetry around the axis perpendicular to the page plane, and Fig. 7d has threefold rotational symmetry around the axis perpendicular to one of the triangular planes of tetrahedra, so they do not have SOS with $\mathcal{M}$. However, these threefold rotational symmetries are broken in the presence of the green-double-arrow strain, so now all of them have SOS with $\mathcal{M}$. These strain-induced SOS relationships with $\mathcal{M}$ indicate that they are a kind of piezomagnets. Note that the two-in-one-out (or one-in-two-out) spin arrangement in a triangle, which can be a part of antiferromagnetic kagome lattice, and the three-in-one-out (or one-in-three-out) spin arrangement in a tetrahedron, which can be a part of antiferromagnetic pyrochlore lattice, in Fig. 7e do have SOS with $\mathbf{P}$ with broken $\{\mathbf{R}, \mathbf{I}, \mathbf{M}\}$ ^{54,55}. In fact, this SOS relationship with $\mathbf{P}$ holds on a triangular lattice as long as two bonds have 60° spin order and one bond with 120° spin order, as shown in Fig. 7e. Also note that when antiferromagnetic states with no external perturbations having SOS with $\mathcal{M}$ tend to, in fact, show tiny net magnetic moments (on the order of $0.001 \mu\text{B}/\text{magnetic ion}$), possibly due to the contribution of orbital magnetic moment, originating from the SOS relationship with $\mathcal{M}$ ⁴⁵. Figure 7f, g depict particular types of planar antiferromagnetic order in AB-stacked triangular lattices, having SOS with $\mathcal{M}$ ^{56,57}. This magnetic lattice is common in hcp systems; however, these types of planar antiferromagnetic order have never been observed experimentally or discussed theoretically, as far as the author is aware of. Anyhow, if these inversion-symmetric antiferromagnetic states are realized, then they will exhibit MOKE, Faraday effect and also anomalous Hall-type effects.

We would like to clarify some of confusing and conflicting issues in literatures. Numerous different kinds of non-collinear and co-planar antiferromagnetic order have been realized in kagome lattice, and many of them have not-broken $\mathbf{R}$ (or C_3) symmetry, so cannot have SOS with $\mathcal{M}$. In addition, some of them (e.g., the Fig. 7b case without green double arrow (i.e. strain) and the 3rd case without green double arrow in Fig. 7c) do have broken $\mathbf{R}$ symmetry, but also maintain threefold rotational symmetry around the out-of-page-plane axis, so cannot have SOS with $\mathcal{M}$. The original prediction of the presence of anomalous Hall effect and MOKE in non-collinear antiferromagnets through a Berry curvature mechanism was done on Mn_3Ir and $\text{Mn}_3(\text{Rh}, \text{Ir}, \text{Pt})$, respectively, with the magnetic state of the 3rd case of Fig. 7c without green double arrow^{44,46,58,59}. However, this magnetic state has threefold rotational symmetry, so cannot have SOS with $\mathcal{M}$. Indeed, the experimental observation of anomalous Hall effect and MOKE (and anomalous Nernst and thermal Hall effects) in non-collinear antiferromagnets was made on Mn_3Sn , Mn_3Ge , and Mn_3Ga ^{45,47–51,60–62}. The magnetic states of Mn_3Sn and Mn_3Ge (Ga) turn out to be the 1st and 2nd cases of Fig. 7a, respectively. Both cases of Fig. 7a do have SOS with $\mathcal{M}$, but the directions of the relevant $\mathcal{M}$'s are along different directions with respect to the underlying kagome lattice.

CONCLUSION

In general, the SOS approach can be applied to numerous physical phenomena in solids such as nonreciprocity, magnetism-induced ferroelectricity, linear magnetoelectricity, optical activities (including MOKE and Faraday effect), photogalvanic effects, second harmonic generation, and anomalous Hall-type effects, and can be leveraged to identify new materials that potentially exhibit the desired physical phenomena. We have proven two relevant and important theorems: (1) Any materials with broken $\{\mathbf{R}, \mathbf{M}, \mathbf{T}\}$, but not-broken $\{\mathbf{R}, \mathbf{I}\}$ (i.e., having SOS with $\mathcal{M}$) can show all of anomalous Hall, anomalous Ettingshausen, anomalous Nernst, and anomalous thermal Hall effects that are linear with $\mathbf{E}_{\text{ext}}$, and (2) all materials having SOS with $\mathcal{M}$ exhibit both Faraday effect and MOKE, and any inversion-symmetric materials exhibiting both Faraday and MOKE do show SOS with $\mathcal{M}$. In this paper, we have deliberated a large number of specimen constituents having SOS with $\mathcal{M}$. Some of these do exhibit a significant measurable magnetization, and others exhibit ferromagnet-like behaviors without any significant magnetization. The former includes linear magnetoelectrics in the presence of $\mathbf{E}$, electric current flow in chiral materials, and moving Neel- or Bloch-type ferroelectric walls. The latter includes particular types of antiferromagnetic states exhibiting various ferromagnet-like behaviors such as MOKE, Faraday effect, and anomalous Hall-type effects. Some other types of antiferromagnetic states with zero net magnetic moment in, e.g., Cr_2O_3 can show MOKE without any Faraday effect nor anomalous Hall-type effects. We have presented numerous new cases for these three categories within the concept of SOS – new theoretically and experimentally. Certainly, these propositions need to be experimentally confirmed, and also the potential microscopic mechanisms for these new phenomena ought to be theoretically explored.

METHODS

The relationships between specimen constituents (i.e., lattice distortions or spin arrangements in external fields or other environments, etc., and also their time evolution) and measuring probes/quantities (i.e., propagating light, electrons, or other particles in various polarization states, including light or electrons with spin or orbital angular momentum, bulk polarization or magnetization, etc., and also experimental setups to measure, e.g., Hall-type effects) are analyzed in terms of the characteristics under various symmetry operations of rotation, space inversion, mirror reflection, and time reversal. When specimen constituents and measuring probes/quantities share the same broken symmetries, except translation symmetry, they are said to exhibit symmetry operation similarities (SOS), and the corresponding phenomena can occur. In terms of SOS, we have considered the specimen constituents for linear magneto-electric effects, production of effective magnetic fields with moving ferroelectric walls, and also production of net magnetization from antiferromagnetic states. In addition, we also discuss the requirements for the observation of MOKE, Faraday-type effects, and/or anomalous Hall-type effects in terms of broken symmetries.

DATA AVAILABILITY

The datasets generated during the current study are available from the corresponding author on reasonable request.

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AUTHOR CONTRIBUTIONS

S.W.C. is solely responsible for obtaining the results and writing the paper.

COMPETING INTERESTS

The author declares no competing interests.

ADDITIONAL INFORMATION

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