

Circular Dichroism in Resonant Inelastic X-Ray Scattering: Probing Altermagnetic Domains in MnTe

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X-ray magnetic circular dichroism provides a means to identify ferromagnetic, chiral, and altermagnetic orders via their time-reversal-symmetry ($\mathcal{T}$) breaking. However, the symmetry properties that govern circular dichroism (CD) in resonant inelastic x-ray scattering (RIXS) remain poorly understood. We show that, due to the inherent irreversibility of the RIXS process, RIXS-CD does not require $\mathcal{T}$ breaking to be present, but reflects the change in unitary symmetries associated with magnetic ordering. Using the altermagnetic MnTe as a model system, we observe an azimuthal-angle dependent RIXS-CD signal in the magnon excitations. Our findings highlight the sensitivity of RIXS-CD to the relativistic symmetry in magnetic systems and its potential application as a probe of magnetic domains.

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The interaction of circularly polarized photons with matter provides a fundamental probe of symmetry-breaking phenomena in solids. Left- and right-circularly polarized photons carry opposite helicities of spin angular momentum along their propagation direction. When they interact with a material, the corresponding transition amplitudes may differ depending on the spontaneously broken symmetries of the system. Circular dichroism (CD) in x-ray absorption has been extensively used to detect time-reversal symmetry ($\mathcal{T}$) breaking in ferromagnets [1,2] and, more recently, in some compensated magnets [3–5]. In the dipole approximation, CD in absorption is strictly forbidden by $\mathcal{T}$, meaning that a finite CD signal serves as a direct experimental signature of $\mathcal{T}$ breaking in magnetic systems. Beyond time-reversal symmetry, circularly polarized photons are also sensitive to mirror symmetry operations, making CD a valuable tool for probing crystallographic symmetry. Recently, CD in

inelastic photon scattering has been utilized to detect chiral phonons [6,7], demonstrating its broader applicability in studying emergent symmetry-driven excitations.

Resonant inelastic x-ray scattering (RIXS) is a rapidly advancing technique that utilizes x-ray photons [8–10]. Unlike x-ray absorption, RIXS provides momentum resolution and is less constrained by spin conservation and dipole selection rules [11,12]. While the linear polarizations of the incoming light have been widely exploited to differentiate charge and spin excitations, the exploration of CD in RIXS (RIXS-CD) for magnetic materials remains largely uncharted. An unrecognized yet crucial property is the *non-Hermiticity* of the RIXS process, discussed later, which inherently breaks $\mathcal{T}$. Thus, RIXS-CD does not require $\mathcal{T}$ breaking, and nonzero RIXS-CD can exist even without spontaneous symmetry breaking (hereafter normal states). Yet, $\mathcal{T}$ breaking by magnetic ordering affects their mirror symmetry properties. With a careful choice of the polarizations and propagation directions of the two photons, RIXS-CD offers unique sensitivity to relativistic symmetry in magnetic systems, which we use here to study altermagnetism.

Altermagnets have emerged as a distinct subclass of collinear magnets, setting themselves apart from antiferromagnets [13–15]. Altermagnets are collinear magnets with zero net magnetization, where the antiferromagnetic

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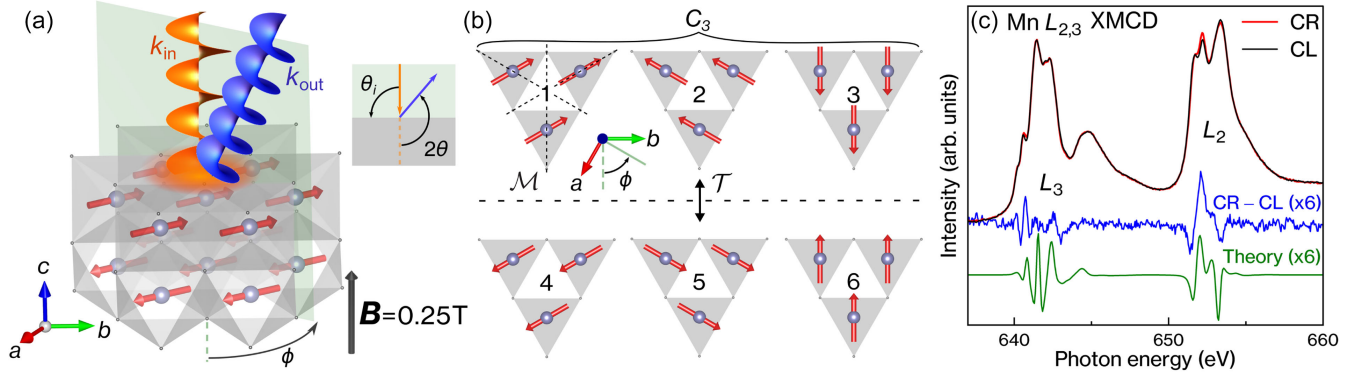


FIG. 1. (a) Altermagnetic order in MnTe and the scattering geometry of the resonant inelastic x-ray scattering (RIXS) experiment. The orange (blue) spiral depicts the incoming (outgoing) circularly polarized x-ray photons. θ_i denotes the angle between the incoming beam and the sample surface, 2θ is the scattering angle, and the azimuthal angle ϕ generates the rotation of the scattering plane around the sample normal. A small magnetic field of $B = 0.25$ T is applied along the c axis to stabilize the depicted altermagnetic domain. (b) Possible six altermagnetic domains. Domains 1–3 are $\mathcal{T}$ -equivalent domains related by the C_3 rotation, and domains 4–6 are their $\mathcal{T}$ counterparts. The dotted lines on domain 1 indicate the mirror planes ($\mathcal{M}$) of the normal state. (c) Mn $L_{2,3}$ -edge x-ray absorption (XAS) spectra collected with total fluorescence yield (TFY) mode using the right (CR) and left (CL) circularly polarized light. The blue curve shows the x-ray magnetic circular dichroism (XMCD) signal. The green curve shows the theoretical XMCD spectrum [5].

sublattices are connected by proper or improper rotations rather than translation or inversion. Under these conditions, the magnetic ground states related by time-reversal operation become macroscopically inequivalent, giving rise to unique properties such as alternating spin polarization in electronic bands [16,17], the anomalous Hall effect [18], and spin currents [19].

X-ray magnetic circular dichroism (XMCD) has proven effective for detecting $\mathcal{T}$ breaking [5] and determining the orientation of the Néel vector $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$ in some altermagnets [20,21], where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations. However, in the archetypal altermagnet α -MnTe (MnTe), where the two Mn ions are related by a screw operation [Fig. 1(a)], XMCD cannot distinguish three magnetic domains related by the C_3 rotation symmetry [Fig. 1(b)]. A way to overcome this difficulty is to combine XMCD with linear dichroism (LD) measurements [22]. On the other hand, the scattering plane in RIXS-CD naturally distinguishes the C_3 domains from the spectral weight of the magnetic excitations.

In this Letter, we investigate Mn L_3 -edge RIXS-CD in the altermagnetic state of MnTe through a combination of measurements and theoretical simulations. We observe a significant RIXS-CD signal in the magnon excitations, which aligns well with our numerical calculations. Interestingly, we find that RIXS-CD is not a consequence of $\mathcal{T}$ breaking. Instead, it reflects the breaking of certain mirror symmetries induced by the $\mathcal{T}$ -broken altermagnetic order, enabling the distinction of magnetic domains in MnTe from the azimuthal angle dependence of the RIXS-CD signal.

Experiment—Mn L_3 -edge (~ 640 eV) RIXS experiment of MnTe single crystals was performed at the I21 beamline [23] of Diamond Light Source. Figure 1(a) depicts the

scattering geometry of the RIXS experiment. θ_i defines the angle between the incoming beam and the sample surface, and the scattering angle 2θ was continuously varied to precisely define the momentum transfer $\mathbf{q} = \mathbf{k}_{\text{in}} - \mathbf{k}_{\text{out}}$ in the three-dimensional reciprocal space. The azimuthal angle ϕ was used to change the in-plane $\mathbf{q}$ path. All the measurements were performed at $T = 24$ K, well below the Néel temperature of 307 K [24]. We applied a small magnetic field of $B = 0.25$ T along the c axis by mounting the single crystal onto a permanent magnet, to stabilize the equivalent $\mathcal{T}$ domains [1–3 in Fig. 1(b)] by utilizing the small canted magnetic moment along the c axis [24]. The total energy resolution of the spectrometer was set to 24 meV. The polarization of the outgoing photons was not analyzed. The momentum transfer $\mathbf{q} = (H, K, L)$ is expressed in the reciprocal lattice units (r.l.u.). The details of the characterization of the MnTe single crystals are provided in Supplemental Material [25].

Theory—We simulate RIXS spectra using two complementary approaches: (i) an atomic calculation combined with the spin-wave theory of the Heisenberg model [27,28], and (ii) the DFT + DMFT Anderson impurity model (AIM) method [11,29,30]. The first approach captures the interference between excitations at different Mn sites, thereby accounting for the $\mathbf{q}$ dependence of the final states. However, it is limited to magnon excitations, and the time evolution of the intermediate state is constrained to a single atom. The second method neglects the momentum dependence of the final states; the total intensity is treated as the sum of contributions from the individual Mn sites [5,31]. Nevertheless, it captures the evolution of the intermediate states and includes all types of excitations. In the first approach, the outgoing momentum determines the final state momentum and constrains the polarization of the

outgoing photon. In the second approach, the outgoing momentum enters only through the photon polarization.

We first demonstrate a dominant population of the altermagnetic $\mathcal{T}$ domains within the investigated x-ray spot by performing an XMCD measurement in the total fluorescent yield (TFY). Note that the XMCD and RIXS-CD measurements below are performed at the same spot. Figure 1(c) shows the x-ray absorption (XAS) spectra collected using the left (CL, black) and right (CR, red) circularly polarized photons. The XMCD signal (blue), defined as their difference, shows oscillatory behavior in the L_3 and L_2 regions. The XAS and XMCD signals near the L_3 edge are suppressed compared to those near the L_2 edge, because of a stronger self-absorption effect in fluorescence x-ray detection. However, the sequence of minima and maxima of the XMCD spectrum and its amplitude near the L_2 region are in excellent agreement with those of the theoretical spectrum (green), which exhibits oscillations characteristic of the altermagnetism in MnTe with the $[1\bar{1}00]$ orientation of the magnetic moments [5]. Note that this frequency dependence in the XMCD spectrum is distinct from the one originating from the net magnetization due to moment canting [5], confirming that the net magnetic moment remains tiny under a magnetic field of 0.25 T [32]. The XMCD amplitude at the L_2 edge closely matches the theoretical prediction for a single domain sample. This is in contrast to the previous XMCD spectrum of a MnTe thin film collected in the total electron yield [5], whose amplitude was about ten times weaker than the theoretical prediction due to the coexistence of opposite $\mathcal{T}$ domains. Note that the XMCD spectrum alone cannot distinguish the three domains connected by the C_3 rotation [domains 1-3 in Fig. 1(b)]. RIXS-CD, on the other hand, allows the distinction of the C_3 domains, as we will show below.

Having confirmed the altermagnetic order, we now examine the magnon dispersion. Figure 2(a) shows the low-energy region of the RIXS spectra along the $\mathbf{q} = (H, 0, -0.5)$ direction, collected with π -polarized $h\nu = 641.1$ eV photons. One readily identifies the elastic peak, magnon excitation at ~ 32 meV, and a shoulder structure around ~ 60 meV, whose plausible origin is a two-magnon excitation [11]. We have fitted the spectra to three Voigt profiles, and the magnon contribution is highlighted with shaded red curves. In agreement with the former INS results in Refs. [33,34], the magnon energy shows a weak dependence on the in-plane H , reflecting subdominant in-plane exchange interactions. On the other hand, the magnon exhibits significant dispersion along the $\mathbf{q} = (0, 0, L)$ direction [Fig. 2(b)], due to the dominant out-of-plane exchange interactions. The magnon energy is maximal at $L = -0.5$ and shows a downward turn as L approaches 0 or -1 .

To describe the magnon dispersion, we use the following $S = 5/2$ spin Hamiltonian:

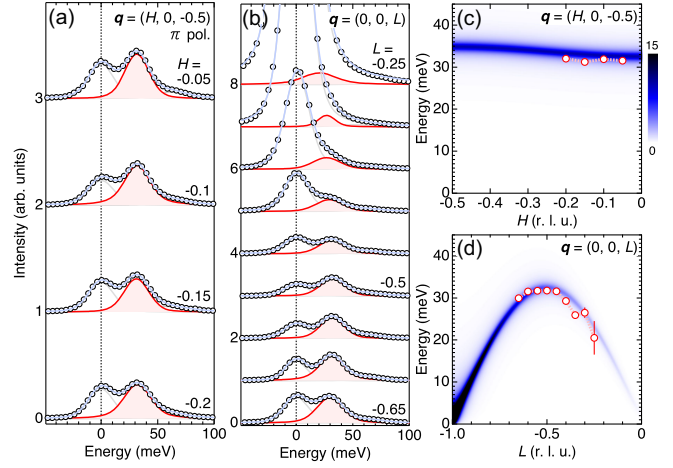


FIG. 2. (a),(b) Low-energy RIXS spectra of MnTe collected along the $\mathbf{q} = (H, 0, -0.5)$ and $\mathbf{q} = (0, 0, L)$ directions. The data were collected with π -polarized photons with $h\nu = 641.1$ eV. Three Voigt profiles represent the elastic, magnon (red), and two-magnon excitations. (c),(d) Transverse component of the spin dynamical structure factor $S(\mathbf{q}, \omega) = (S^{yy} + S^{zz})(\mathbf{q}, \omega)$. The experimental magnon peak energies are plotted as red circles.

$$\mathcal{H} = \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + D \sum_i (S_i^x)^2,$$

where J_{ij} represents the Heisenberg interactions between spins on the i th and j th Mn sites, and D is the single-ion anisotropy term. Figures 2(c) and 2(d) compare the experimental magnon dispersions (red circles) with the transverse component of the spin dynamical structure factor, $S(\mathbf{q}, \omega) = (S^{yy} + S^{zz})(\mathbf{q}, \omega)$, where the x axis is taken parallel to the Néel vector $\mathbf{L}$. We have computed the excitation spectra for the magnetic ground state of domain 1. The optimal parameters, computational methods, and high-energy multiplets that confirm the $S = 5/2$ ground states are detailed in Supplemental Material [25]. $S(\mathbf{q}, \omega)$ along $\mathbf{q} = (H, 0, -0.5)$ in Fig. 2(c) exhibits an almost flat H dependence, reproducing the experimental data within the accessible $\mathbf{q}$ space. Also, $S(\mathbf{q}, \omega)$ along $\mathbf{q} = (0, 0, L)$ in Fig. 2(d) reaches a maximum at $L = -0.5$ and approaches $\omega = 0$ as L approaches 0 or -1 , in agreement with the RIXS data and the INS results [34]. The splitting of the two magnon dispersions along these high-symmetry $\mathbf{q}$ paths is almost negligible except near the bottom of the band, as it is solely caused by the relativistic D term.

Next, we investigate the RIXS-CD at magnon excitations. As depicted in Fig. 1(a), circularly polarized x-ray photons were incident parallel to the c axis ($\theta_i = 90^\circ$), and the outgoing photons were detected at the scattering angle of $2\theta = 150^\circ$. The top panels in Figs. 3(a)–3(c) display the RIXS spectra collected with circularly polarized photons, obtained at three azimuthal angles $\phi = 0^\circ$, 30° , and 60° , respectively. For the real-space definition of $\phi = 0^\circ$, see Fig. 1(b). The RIXS-CD spectra are also

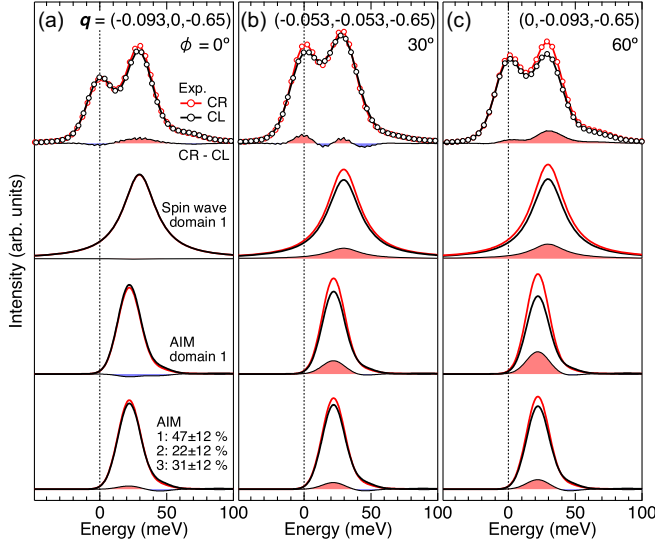


FIG. 3. (a)–(c) RIXS spectra collected with right and left circularly polarized light, at the azimuthal angles $\phi = 0^\circ$, 30° , and 60° , respectively. RIXS-CD intensity defined as their difference is also shown. The curves in the second and third rows are the corresponding theoretical RIXS and RIXS-CD spectra for the domain 1 computed with the spin wave and Anderson impurity model (AIM) methods. The bottom curves show the fit of the experimental curves as superpositions of the theoretical AIM spectra for the domains 1–3.

presented, with positive and negative regions shaded in red and blue, respectively. The momentum transfers are $\mathbf{q} = (-0.093, 0, -0.65)$, $(-0.053, -0.053, -0.65)$, and $(0, -0.093, -0.65)$, all sharing the same out-of-plane component $L = -0.65$ but differing in their projections to the ab -plane. The CD at the magnon peak is clearly observed at $\phi = 60^\circ$, while the CD at $\phi = 0^\circ$ and 30° is weaker.

To understand this ϕ dependence in RIXS-CD, the curves in the second and third rows in Fig. 3 show the theoretical RIXS and RIXS-CD spectra obtained using the spin wave and AIM methods, respectively. The calculations are performed for the single domain 1 in Fig. 1(b). First, we find that both methods yield maximal CD intensities at $\phi = 60^\circ$, and negligibly small CD at $\phi = 0^\circ$. Note that these two directions correspond to the largest amplitude of the g -wave altermagnetism of MnTe with the opposite signs. Correspondingly, the two magnon bands with opposite chiralities show energy splitting in opposite directions. Nevertheless the RIXS-CD intensity does not show a simple sign reversal with the same amplitude in either experiment or theory. This observation, together with the reasonable agreement with impurity calculations that exclude propagating magnons, demonstrates that the RIXS-CD amplitude in MnTe is not directly linked to the magnitude of the g -wave altermagnetic magnon splitting, contrary to the interpretation proposed in Ref. [35]. Instead, RIXS-CD

reflects orientation of the outgoing photon polarization with respect to the $\mathbf{L}$ vector of the ground state. This allows us to use RIXS-CD to distinguish the C_3 domains.

Here, we clarify the origin of CD in the RIXS process. In general, the Kramers-Heisenberg formula for the RIXS cross section $F^\pm(\mathbf{k}_{\text{out}}) \equiv [\delta^2 \sigma_\pm / \delta \Omega \delta \omega](\mathbf{k}_{\text{out}})$ [36],

$$\begin{aligned} F^\pm(\mathbf{k}_{\text{out}}) &\propto \sum_{f,\alpha} \left| \sum_m \frac{\langle f | V_\alpha | m \rangle \langle m | V_\pm | i \rangle}{\omega_{\text{in}} + E_i - E_m + i\Gamma} \right|^2 \\ &\quad \times \delta(\omega_{\text{in}} - \omega_{\text{out}} + E_i - E_f) \\ &\equiv \sum_{f,\alpha} \left| \sum_m \langle f | V_\alpha X V_\pm | i \rangle \right|^2 \delta(\dots), \end{aligned}$$

has the form of the Fermi golden rule connecting the initial $|i\rangle$ and final $|f\rangle$ states with respective energies $E_i + \omega_{\text{in}}$ and $E_f + \omega_{\text{out}}$. Notably, the operator $X = \sum_m [(|m\rangle\langle m|) / \omega_{\text{in}} + E_i - E_m + i\Gamma]$, which describes the propagation in the intermediate state, is non-Hermitian due to the imaginary part of the denominator. The non-Hermiticity arises from the resonant processes, where the initial and final states (including photons) are degenerate with a continuum of intermediate states. This property persists even as $\Gamma \rightarrow 0$. Consequently, unlike XMCD, RIXS is not inherently sensitive to $\mathcal{T}$ breaking, and nonzero CD can exist even in the normal state. However, unitary symmetries, such as crystallographic symmetries, can impose selection rules that suppress RIXS-CD. Since the incoming and outgoing photons are part of the initial and final states ($|i\rangle$ and $|f\rangle$), their momenta ($\mathbf{k}_{\text{in}}$ and $\mathbf{k}_{\text{out}}$) must be included in the symmetry analysis. For instance, RIXS-CD is forbidden if both $\mathbf{k}_{\text{in}}$ and $\mathbf{k}_{\text{out}}$ lie in the same mirror plane. In the present geometry, this symmetry argument leads to the vanishing of RIXS-CD at $\phi = (n \times 60)^\circ$ in the normal state [see the mirror planes ($\mathcal{M}$) in Fig. 1(b)]. Thus, the nonzero RIXS-CD at $\phi = 0^\circ$ and 60° in our experiment is allowed by the mirror-symmetry breaking associated with the altermagnetic order.

In the AIM approximation, the selection rule becomes even more restrictive. The total scattered intensity averaged over outgoing light polarizations is then given by [25]

$$F^\pm(\mathbf{k}_{\text{out}}) \propto \sum_f \left(\text{Tr}(\mathbf{M}_{fi}^\pm \bar{\mathbf{M}}_{fi}^\pm) - \hat{\mathbf{k}} \cdot \mathbf{M}_{fi}^\pm \bar{\mathbf{M}}_{fi}^\pm \cdot \hat{\mathbf{k}} \right),$$

with $\mathbf{M}_{fi}^\pm = \langle f | \mathbf{V} X \mathbf{V}^\pm | i \rangle$ and $\hat{\mathbf{k}} = \mathbf{k}_{\text{out}} / |\mathbf{k}_{\text{out}}|$. This implies the $F_n^\pm(\phi) = A^\pm + B^\pm \cos(2\phi + \frac{2}{3}\pi n + \phi_0^\pm)$ dependence of the RIXS amplitude, where $n = 1, 2, 3$ is the index of the C_3 domain in Fig. 1(b). As a result, an equal domain population leads to a ϕ -constant RIXS-CD signal ($\Delta F_n = F_n^+ - F_n^-$). The mirror-plane relationship between domains 1 and 4 implies $\Delta F_4(\phi) = -\Delta F_1[-\phi - (2\pi/3)]$. The $\mathcal{T}$ domains thus lead to a RIXS-CD signal with

opposite constant part and mirror ϕ dependence. Since the mirror plane is a symmetry of the normal state, the RIXS-CD vanishes for arbitrary ϕ in the present geometry. Nevertheless, for a general k_{in} CD is allowed even in the normal state. For details see Supplemental Material [25].

The strong azimuthal dependence of the experimental RIXS-CD, therefore, points to a considerable difference in the domain populations within the measurement spot. We fitted the experimental RIXS-CD curves to superpositions of the AIM curves of the three domains [1–3 in Fig. 1(b)]. Here, the population ratio was optimized to reproduce the angular dependence of the RIXS-CD amplitude [25]. The fitted curves, shown at the bottom of Figs. 3(a)–3(c), show good agreement with the experimental data for all the three ϕ angles, including the overall positive sign originating from the constant term of $F_n^{\pm}(\phi)$. As expected from the largest CD at $\phi = 60^\circ$, domain 1 has the dominant occupation of $47\% \pm 12\%$, while domains 2 and 3 also have nonnegligible occupations of $22\% \pm 12\%$, and $31\% \pm 12\%$, respectively. The errors represent the standard deviations in the fitting process, which can be reduced by increasing the number of data points [25]. The azimuthal dependence of RIXS-CD thus allows quantitative estimation of magnetic domain weights.

In summary, we have conducted an Mn L_3 RIXS study of the magnetic excitations in the altermagnetic MnTe. The azimuthal dependence of the RIXS-CD intensity reveals a dominant occupation of a single altermagnetic domain. The observed nonzero RIXS-CD in our scattering geometry is attributed to the mirror-symmetry breaking associated with the T -breaking altermagnetic order. Our findings establish RIXS-CD as a domain-sensitive probe of elementary excitations in a broad range of transition metal compounds. RIXS-CD detects the unitary (space) symmetry breaking via T -breaking magnetic orders. This makes RIXS-CD a complementary technique to XMCD, with potential applications to a wider class of anomalous Hall antiferromagnets, including the noncollinear ones [37,38].

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Data availability—The data that support the findings of this article are openly available [39].

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Correction: In the equation for the Kramers-Heisenberg formula for the RIXS cross section, a misplaced superscript has been fixed. A support statement has been added to the Acknowledgments section.