Editors' Suggestion

Magnetoelastic Dynamics of the Spin Jahn-Teller Transition in $CoTi_2O_5$

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$CoTi_2O_5$ has the paradox that low temperature static magnetic order is incompatible with the crystal structure owing to a mirror plane that exactly frustrates magnetic interactions. Despite no observable structural distortion with diffraction, $CoTi_2O_5$ does magnetically order below $T_N \sim 25$ K with the breaking of spin ground state degeneracy proposed to be a realization of the spin Jahn-Teller effect in analogy to the celebrated orbital Jahn-Teller transition. We apply neutron and Raman spectroscopy to study the dynamics of this transition in $CoTi_2O_5$. We find anomalous acoustics associated with a symmetry breaking strain that characterizes the spin Jahn-Teller transition. Crucially, the energy of this phonon coincides with the energy scale of the magnetic excitations, and has the same symmetry of an optic mode, observed with Raman spectroscopy, which atypically softens in energy with decreasing temperature. Taken together, we propose that the energetics of the spin Jahn-Teller effect in $CoTi_2O_5$ are related to cooperative magnetoelastic fluctuations as opposed to conventional soft critical dynamics which typically drive large measurable static displacements.

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Structural phase transitions are typically driven by a dynamic soft mode [1–4] which freezes either continuously or discontinuously. Classic examples include soft zone center transverse optical phonons in perovskites [5–7] and zone boundary phonon anomalies that appear in orientational-like order-disorder transitions [8,9]. Such structural transitions are readily observed through diffraction techniques. However, there is a growing list of materials where a magnetostructural transition must occur based on symmetry constraints or bulk measurements, yet are not accompanied by an observable change in the Bragg peaks measured with x-ray or neutron diffraction. For example, this is the case for ferroaxial multiferroic phase transitions in $RbFe(MoO_4)_2$ [10,11], $CaMn_7O_{12}$ [12,13], and $Cu_3Nb_2O_8$ [14,15], where changes in Bragg peaks and soft lattice dynamics are not readily observable [16]. This is despite known low temperature structural transitions derived based on symmetry constraints. A further example is certain ferroelectric metal-organic frameworks [17] which lack observable soft phonon dynamics despite known displacive ferroelectric transitions.

We address this apparent contradiction through studying the lattice and magnetic dynamics of a similar problem in the spin-Jahn Teller material $CoTi_2O_5$, where a structural transition is not observable in conventional diffraction, yet must occur to allow low temperature magnetic order, which is exactly frustrated based on the high temperature nuclear structure. In analogy to the orbital Jahn-Teller theorem [18] that predicts a structural distortion will occur to break an orbital degeneracy, a similar idea has been proposed in the context of spin degeneracy from frustrated magnetism where an orbital degeneracy is absent. The "spin Jahn-Teller effect" is where a spin degeneracy drives a symmetry breaking structural transition that lifts the degeneracy of the spin manifold and facilitates long-range magnetic order.

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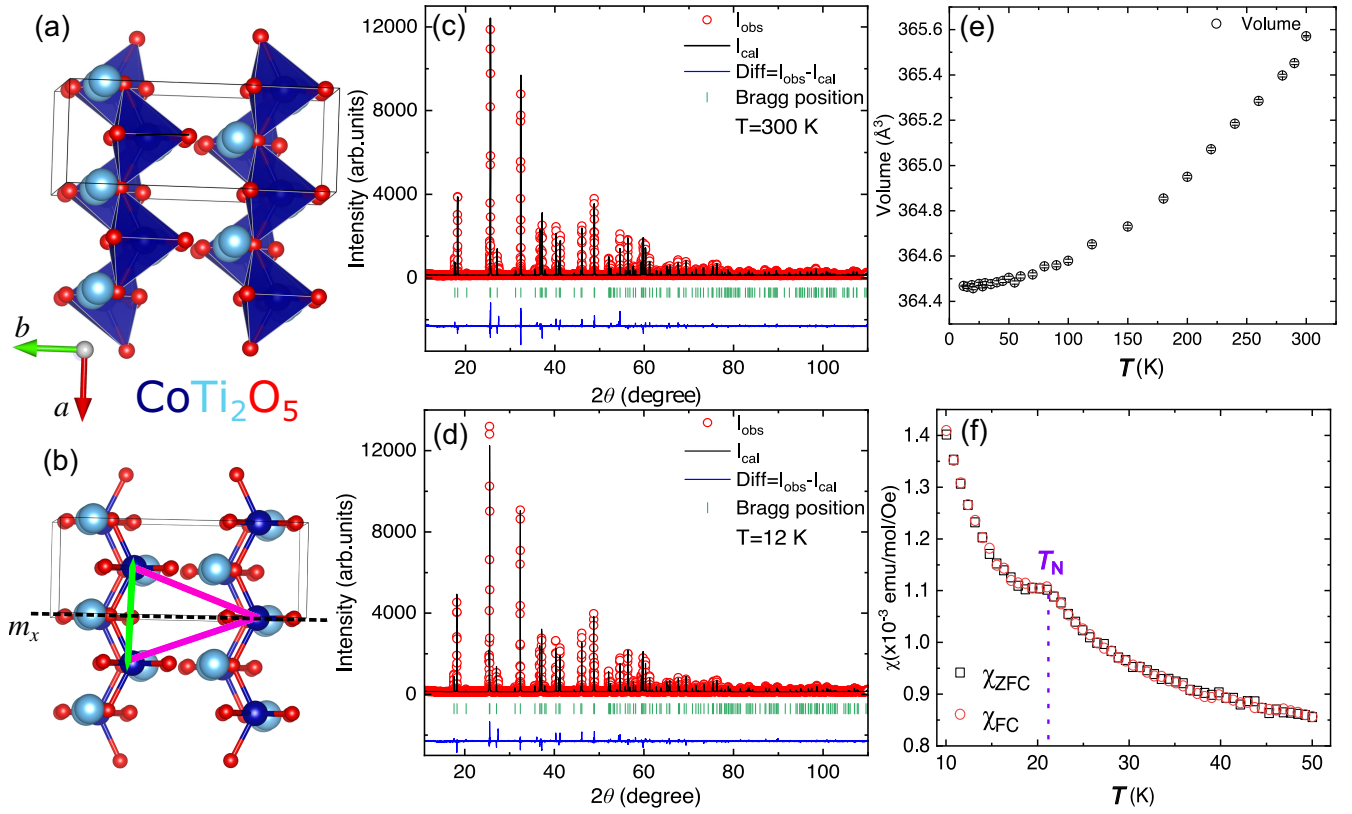

FIG. 1. Structural and magnetic properties of $CoTi_2O_5$. (a),(b) The nuclear structure and the frustrating triangular arrangement of the interactions. (c),(d) The same diffraction measurements at 300 and 12 K. (e) A plot of the volume of the lattice with temperature showing no change in slope at the (f) Néel temperature of ~23 K measured in a powder sample with an applied field of 100 G.

Such a structural distortion originates from the competition between magnetic and elastic energy scales [19,20]. Given the reduction of the magnetic energy scales linearly with atomic positions while the elastic scales quadratically, it is favored for distortion to occur for the relief of frustrating magnetic interactions, resulting in spatially long-range magnetic order. We study the dynamics of this effect in $CoTi_2O_5$ by applying neutron and Raman spectroscopy. While a structural transition is not observable, we find evidence of acoustic dynamics associated with atomic displacement that is linked with magnetic frustration.

$CoTi_2O_5$ adopts a pseudobrookite structure with an orthorhombic space group *Cmcm* at high temperatures [21]. The magnetic Co^{2+} ions reside on a site with $m2m(C_{2v})$ symmetry, implying that the orbital levels are already nondegenerate and therefore the normal Jahn-Teller theorem [22] discussed above does not apply. The lack of an orbital degeneracy in $CoTi_2O_5$ is further discussed in the Supplemental Material [23] in the context of the search for spin-orbit transitions seen in octahedrally coordinated and orbitally active Co^{2+} based compounds [24–28] which we find are absent in $CoTi_2O_5$. The frustrated antiferromagnetic interactions, mediated by indirect exchange through oxygen, are illustrated in Figs. 1(a) and 1(b) and are based on exactly isosceles triangles. Because of a crystallographic mirror plane perpendicular to [100] (termed m_x), this is a perfectly frustrated geometry resulting in spin order that is expected to be unstable to thermal fluctuations. Therefore, magnetic order is not expected unless accompanied by a structural distortion (displacive and strain) [29] that transforms as the Γ_2^+ irreducible representation as identified in Ref. [30]. However, as shown in Ref. [30] applying high resolution neutron diffraction, $CoTi_2O_5$ does not undergo an observable structural transition yet displays spatially long-range magnetic order below $T_N \sim 25$ K. It was therefore concluded that $CoTi_2O_5$ undergoes a spin Jahn-Teller transition where the structure distorts, subtly, to break the large ground state spin degeneracy imposed by the frustrating geometry. Further studies have been performed on isostructural $FeTi_2O_5$ and have also found similar results [31]. In the following, we investigate acoustic and optic structural dynamics near this spin Jahn-Teller transition and probe low-energy magnetic excitations that characterize low temperature magnetic order.

Neutron spectroscopy was used to study the low-energy acoustic phonons and magnetic excitations. Acoustic phonon measurements were performed on the EIGER thermal triple-axis spectrometer (PSI). High resolution magnetic spectroscopy was performed on the IN12 cold-triple axis

spectrometer (ILL). For all neutron scattering experiments, the sample was aligned such that reflections of the form (H, K,0) were within the horizontal plane. We discuss the magnetic dispersion along the (0,0,L) direction in Supplemental Material [23] applying the time of flight MAPS spectrometer. The sample was grown using the traveling floating zone technique [30]. Monochromatic x-ray diffraction as a function of temperature was carried out on a Rigaku Smartlab with a Johansson monochromator and PheniX displex. Temperature dependent Raman spectroscopy was performed to study optical phonons. Further experimental information is in Supplemental Material [23].

Figure 1 reviews the static structural and magnetic properties of $CoTi_2O_5$. The nuclear structure summarized in Figs. 1(a) and 1(b) is built on chains of Co^{2+} ions along the crystallographic a axis. The chains are coupled along the crystallographic b axis via an isosceles triangular arrangement which is exactly frustrated. Monochromatic x-ray diffraction at 300 K Fig. 1(c) is compared to 12 K Fig. 1(d) with no structural distortion observed between these two temperatures (refinement and analysis discussed in Supplemental Material [23] [32,33]). This is further confirmed in Fig. 1(e), where we plot the unit cell volume as a function of temperature. At low temperatures, there is no measurable change in slope which if observed would indicate a structural distortion at T_N. However, as illustrated in Fig. 1(f), despite the frustrating geometry of the spins, a magnetic transition occurs at $T_N \sim 23$ K evidenced by a peak in the magnetic susceptibility.

We now discuss the low-energy magnetic excitations below T_N. Energy-momentum slices along K and H are shown in Figs. 2(b), 2(c), and 2(d), respectively, illustrating energetically gapped [34] magnetic excitations. Confirming expectations based on bonding in the structure and the frustrating triangular arrangement outlined above, the excitations are dispersionless along the K [Fig. 2(b)] direction and strongly dispersive along H [Figs. 2(c) and 2(d)] where Co^{2+} ions are arranged in one-dimensional chains with strong interactions along the crystallographic a axis. We note that the scans along K display two distinct magnetic modes, as expected given the presence of two magnetic ions per primitive unit cell. Fits to the low energy magnetic dispersion applying a nearest-neighbor anisotropic one-dimensional model give a coupling of 3.3 ± 0.2 meV along the chain direction, an anisotropy $D = 0.25 \pm 0.12$ meV, and less than 0.07 meV along b defined by the experimental resolution on IN12. Further measurements discussed in the Supplemental Material [23] indicate a weak 0.3 ± 0.15 meV exchange along c. The lack of any observable dispersion along the K axis in Fig. 2(b) is consistent with no measurable magnetic exchange along the crystallographic b axis. This is expected given the frustrating geometry imposed by the m_x mirror plane; however, is unexpected given the presence of low-temperature spatially long-range magnetic order.

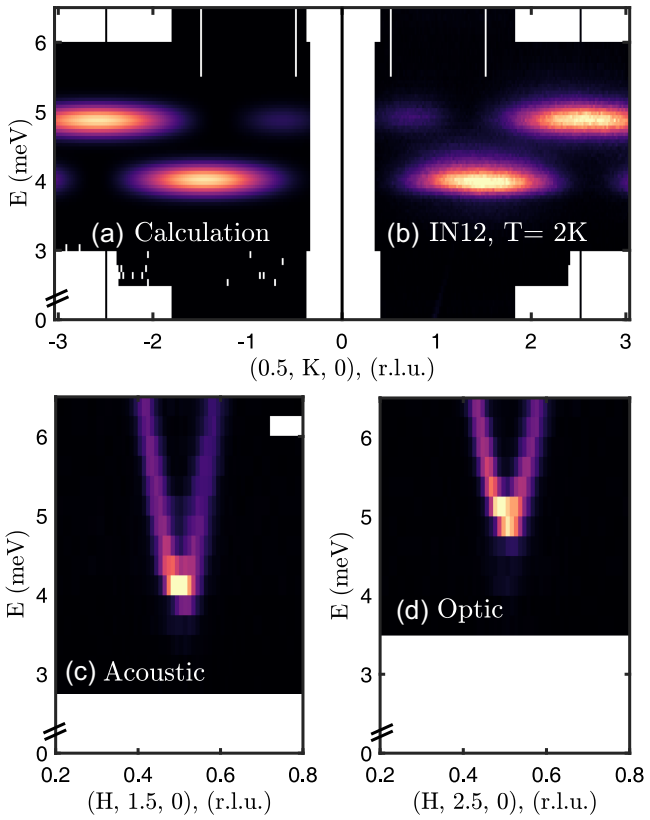

FIG. 2. The low-energy magnetic excitations in the magnetically ordered phase. Given there are two magnetic ions in the unit, there are two spin-wave branches which disperse along (H,0,L) with differing phase, termed acoustic and optic here. (a) Calculated structure factors and (b) measured magnetic correlations perpendicular to the chain axis illustrating nonmeasurable dispersion indicating weak coupling. This contrasts with the resulting coupling along the chain direction displayed in (c), (d) for each of the two magnetic domains.

To understand the intensity modulation along K we compare the data to the nearest neighbor "buckled sheet" model (Supplemental Material [23], Fig. 5). Considering two Co^{2+} sites in a given a–b plane, the neutron scattering intensity from optic and acoustic correlations would take the form of $I(K) \propto [1 \pm \cos(4\pi K y)]$, with $y \sim 0.1911$. The $\pm$ sign fixes the relative excitation phase for the two antiferromagnetic magnon modes in the (H, L) plane with $-$ corresponding to acousticlike (in-phase) magnetic fluctuations and $+$ being opticlike with out-of-phase-like fluctuations of the neighboring spins. Figure 2(a) is a calculation to this model fixing spin excitations at 4 and 4.85 meV. Dispersive H slices for these two modes are shown in Figs. 2(c) and 2(d) (see Supplemental Material [23] for further details).

With a crystallographic distortion not observable with high resolution x-ray or neutron diffraction and no measurable dispersion of the spin excitations along b, we investigate the acoustic shear phonons in Fig. 3. Acoustic lattice fluctuations can be very sensitive to small structural

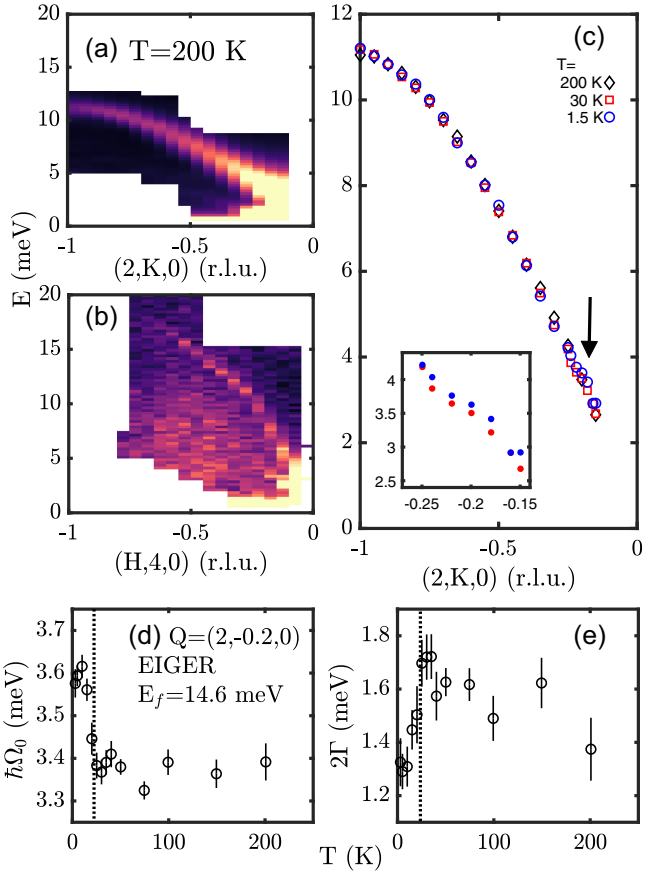

FIG. 3. Acoustic phonons in $CoTi_2O_5$ with the dispersion at $T = 200$ K for acoustic phonons propagating along b^* (a) and a^* (b) (note the momentum and energy broadened paramagnetic scattering near $H \sim -0.5$). An anomaly in the dispersion is evident at $\vec{Q} \sim (2, -0.2, 0)$ (c). (d) and (e) The energy position ($\hbar\Omega_0$) and full width at half maximum (2Γ) as a function of temperature.

distortions as motivated by recent studies linking low-energy acoustic phonons with weak nematic order in pnictides and chalcogenides [35,36], previous works on the dynamic Jahn-Teller effects in rare earth compounds [37–39], and also the sensitivity of acoustic phonons to orbital Jahn-Teller effects [40–46]. Figure 3(a) displays the shear mode with Γ_2^+ symmetry (B_{1g} in the notation of Ref. [47]) and Fig. 3(b) the shear mode with Γ_4^+ symmetry (B_{2g} in Ref. [47] notation). The Γ_2^+ shear mode [Fig. 3(a)] has a lower energy and is defined by a shear with unique crystallographic c axis with the correct symmetry to break the m_x mirror plane in Fig. 1(b). Given its lower energy scale and correct symmetry, we study the temperature dependence of this acoustic phonon in Fig. 3(c), noting a small anomaly in the dispersion at $K \sim -0.2$ displayed in the inset.

Figures 3(d) and 3(e) plot the energy values ($\hbar\Omega_0$) and full width (2Γ) against temperature obtained from constant $\vec{Q} = (2, -0.2, 0)$ scans fit to a damped harmonic oscillator convolved with the experimental resolution (discussed in Supplemental Material [23]). The measured Néel transition temperature, from susceptibility, is marked by the dashed line in both panels. On cooling below T_N, there is an abrupt hardening of the acoustic phonon at this position accompanied by a sharpening in energy indicative of increased phonon lifetime. Given that in the limit $q \to 0$ the acoustic phonon velocity is related [48] to the elastic constant (in this case C_{66} [47]), the hardening of this acoustic phonon at low temperatures in the magnetically ordered phase is indicative of a dynamic strain. We suggest this is a dynamic effect as no change on this scale (5%–6%) is observed in the cell volume [49] presented in Fig. 1 or splitting of nuclear Bragg peaks on the scale reported for similar energy shifts in the acoustic phonons in other materials [50].

Having observed an anomaly in the acoustic phonons at T_N with the correct symmetry to break the frustrating m_x mirror plane, we now study the optical phonons with a single crystal sample using Raman spectroscopy. $CoTi_2O_5$ has 16 atoms in the primitive unit cell, giving 48 positional degrees of freedom. The Γ point, 48-dimensional atomic displacement representation, Γ_r, decomposes into the following irreducible representations; $[\Gamma_r = 8\Gamma_1^+ + 5\Gamma_2^+ + 8\Gamma_3^+ + 3\Gamma_4^+ + 3\Gamma_1^- + 8\Gamma_2^- + 5\Gamma_3^- + 8\Gamma_4^-]$. Of the 45 optic modes, 24 are Raman active (those that are parity even, as denoted by a $+$ subscript). Polarized Raman spectroscopy was performed in reflection geometry, with incident and scattered wave vectors parallel to the c axis (Z). In this geometry only the 8 Γ_1^+ and 5 Γ_2^+ modes are excited. Furthermore, the Γ_1^+ excitations are isolated in the unrotated polarization channels $-Z(XX)Z$ and $-Z(YY)Z$ (in Porto's notation, where $X||a$ and $Y||b$), and the Γ_2^+ excitations are isolated in the rotated $-Z(XY)Z$ and $-Z(YX)Z$ channels.

Figure 4(a) shows the Raman spectra displaying all 5 Γ_2^+ modes with the same symmetry as the anomalous acoustic mode (related to C_{66}) discussed above. The temperature dependence of select modes is presented in Figs. 4(b)–4(d). Arguably, all Γ_2^+ modes display an anomaly at T_N, however, we note these effects are less than 0.025 meV which is negligible in comparison to the energetics of the Γ_2^+ acoustic phonon and magnons discussed above. Figure 4(c) illustrates a softening with temperature of the phonon mode near 21.5 meV that contrasts with the hardening typically expected with decreasing temperature and observed for the other optical phonons (see Supplemental Material [23] for temperature dependence of all 5 Γ_2^+ modes). Phonons typically harden in energy owing to loss of anharmonic effects and lattice contraction [Fig. 1(e)], and energetic softening supports a structural instability with the same symmetry analogous to soft optic phonon driven transitions in classic perovskites [3,5]. This supports a structural instability at T_N with Γ_2^+ symmetry

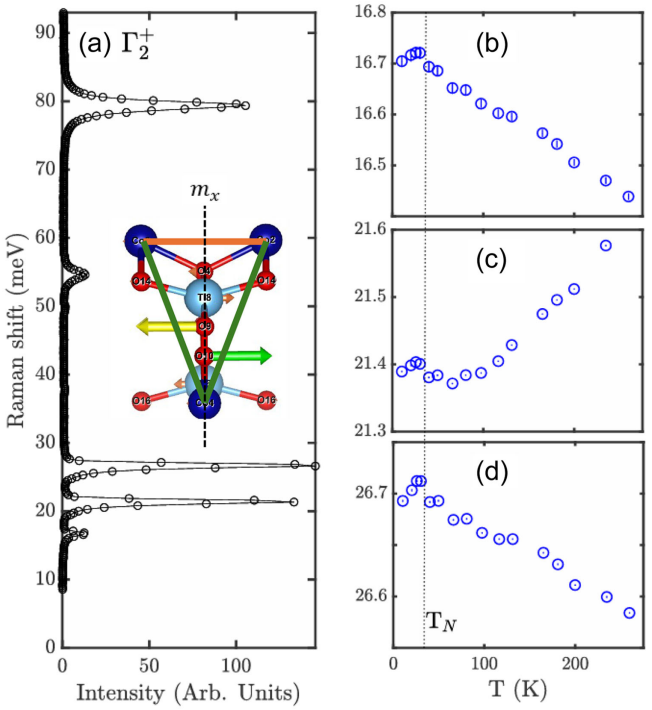

FIG. 4. (a) Raman spectra sensitive to optic phonon modes with Γ_2^+ symmetry which break the frustrating m_x mirror plane illustrated in the inset through the motion of the oxygen atoms. (b)–(d) Temperature dependence of several modes with (c) displaying a softening.

required to break the magnetic frustration outlined in Fig. 1(b).

Density functional theory was used to calculate the phonon eigenvectors and energies which were in agreement with the observed phonon mode energies (see Supplemental Material [23]) and [51–54] for parameter choices. We highlight in the inset of Fig. 4(a) the atomic displacements (eigenmode) associated with the ~21.5 meV mode displayed in Fig. 4(c). The displacements tied with this particular optic phonon mode move the oxygen atoms out of the mirror plane m_x. This breaks the frustration illustrated in Figs. 1(a) and 1(b). We note that the low-energy acoustic shear mode, presented above (Fig. 3), couples directly to the optic mode as it has the same Γ_2^+ symmetry [47] and importantly has the same energy as the dispersionless magnetic excitations along K. This is suggestive of magnetoelastic coupling confirmed through recent macroscopic strain measurements that are able to switch magnetic domains in this compound [55].

We observe an anomalous temperature dependence at T_N of an acoustic phonon mode with the correct symmetry to break the frustrating magnetic interactions [illustrated in Figs. 1(a) and 1(b)] in the absence of an observable structural distortion with diffraction. The energy scale of this anomaly [Fig. 3(c)] is comparable to the energy scale of the magnetic exchange $J \sim 3$ meV and T_N, while significantly larger than the crystalline anisotropy $D \sim 0.25$ meV discussed above. This indicates a coupling between strain and magnetic order. Supporting this and as noted in Ref. [55], considering structural and magnetic order parameters (defined by δ and Φ, respectively), the lowest order invariant in the Landau expansion scales as $\sim\delta\Phi^2$ illustrating a phenomenological mechanism for coupling strain and magnetism in $CoTi_2O_5$. Based on experimental and symmetry grounds, we propose that the magnetic transition in $CoTi_2O_5$ is dynamically driven through cooperative magnetoelastic fluctuations rather than conventional soft phonon dynamics (like in perovskites) or more exotic mechanisms such as nematic order or biquadratic exchange suggested in other triangular magnets [56–58].

In summary, we report neutron inelastic scattering results that reveal a Γ_2^+ shear phonon that hardens at T_N. Furthermore, this phonon mode has the same symmetry as an anomalous optic phonon probed with Raman spectroscopy. The Γ_2^+ acoustic shear mode energetics (in Fig. 3) coincide with those of the magnons presented in Fig. 2. The absence of a measurable static structural distortion nor a magnetic dispersion along K appears to discount a conventional magnetostructural displacive phase transition that breaks m_x. Instead, we propose that our characterization of the dynamics implies that spin Jahn-Teller physics is driven by magnetoelastic dynamics. We note that our results indicate the role of acoustic phonons in establishing the required magnetostructural coupling is at the heart of spin Jahn-Teller physics, when the acoustic phonon and magnon energetics are comparable and of the same symmetry. Our results also suggest that acoustics are a promising avenue to investigate for the study of critical dynamics of magnetostructural transitions in materials where conventional soft lattice dynamics and changes in Bragg peaks are not observable.

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