





Elastic softness of low-symmetry frustrated ATi_2O_5 ($A = Co, Fe$)Tadataka Watanabe ^{1,*}, Kazuya Takayanagi,¹ Ray Nishimura,¹ Yoshiaki Hara,² Dharmalingam Prabhakaran,³ Roger D. Johnson ⁴ and Stephen J. Blundell ³¹*Department of Physics, College of Science and Technology, Nihon University, Chiyoda, Tokyo 101-8308, Japan*²*National Institute of Technology, Ibaraki College, Hitachinaka 312-8508, Japan*³*Department of Physics, Clarendon Laboratory, Oxford University, Parks Road, Oxford OX1 3PU, United Kingdom*⁴*Department of Physics and Astronomy and London Centre for Nanotechnology, University College London, London WC1E 6BT, United Kingdom* (Received 7 August 2024; revised 12 December 2024; accepted 23 January 2025; published 30 January 2025)

Orthorhombic pseudobrookites $CoTi_2O_5$ and $FeTi_2O_5$ have a low-symmetry crystal structure comprising magnetic Co^{2+}/Fe^{2+} ions and nonmagnetic Ti^{4+} ions, where the orbital-nondegenerate Co^{2+}/Fe^{2+} ions form one-dimensional chains running along the orthorhombic a axis. These compounds undergo an antiferromagnetic phase transition at $T_N \sim 26$ K for $CoTi_2O_5$ and $T_N \sim 40$ K for $FeTi_2O_5$. Ultrasound velocity measurements on single crystals of $CoTi_2O_5$ and $FeTi_2O_5$ reveal that $CoTi_2O_5$ exhibits unusual elastic softness above T_N in the symmetry-lowering elastic mode of ac -plane shear elastic modulus, inconsistent with the structural symmetry breaking caused by antiferromagnetic ordering at T_N . This suggests the presence of two distinct types of magnetostructural fluctuations above T_N that should be a precursor to the symmetry-lowering lattice distortion at T_N . In contrast, $FeTi_2O_5$ exhibits either negligible or smaller elastic softness, indicating weaker spin-lattice coupling. These findings highlight $CoTi_2O_5$ and $FeTi_2O_5$ as unique spin-lattice-coupled frustrated systems with low crystal symmetry, where, while the exchange interactions are quasi-one-dimensional in nature, the frustration is released by further lowering the crystal symmetry through three-dimensional spin-lattice coupling, which is stronger in $CoTi_2O_5$ than in $FeTi_2O_5$.

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I. INTRODUCTION

Frustrated magnets provide fertile ground for exploring novel correlated phenomena emerging from competing magnetic interactions [1]. In a frustrated system with spin-lattice coupling, the spin degeneracy can be lifted by symmetry-lowering lattice distortion [2,3]. This effect is called the spin Jahn-Teller (spin-JT) effect because its mechanism is analogous to the Jahn-Teller effect in orbital-degenerate systems where the spontaneous lattice distortion reduces the crystal symmetry to lift the orbital degeneracy [4]. The spin-JT effect was initially proposed for cubic pyrochlore antiferromagnets where spins form a lattice of corner-sharing tetrahedra [2,3]. For real pyrochlore antiferromagnets, the spin-JT mechanism has been applied to explain the magnetostructural transitions of some cubic spinels, where a cubic-to-tetragonal lattice distortion releases the frustration [5–7].

Recently, orthorhombic pseudobrookites ATi_2O_5 ($A = Co, Fe$), having much lower symmetry than the prototypical spin-JT system of cubic spinels, have been proposed as candidate spin-JT systems [8,9]. These pseudobrookites comprise magnetic Co^{2+} ($3d^7$, $S = 3/2$) or Fe^{2+} ($3d^6$, $S = 2$) ions and nonmagnetic Ti^{4+} ions. In addition, the magnetic A^{2+} ions occupying the equivalent crystal sites form one-dimensional (1D) chains running along the orthorhombic a axis (Fig. 1). In ATi_2O_5 , the magnetic A^{2+} ions with C_{2v}

site symmetry have no orbital degeneracy [Fig. 1(*)], and the interchain exchange interactions are considered to be frustrated [8,9]. For $FeTi_2O_5$, the absence of orbital degeneracy was confirmed by Wannier function projection of nonmagnetic generalized-gradient-approximation calculations for the experimental crystal structure [Fig. 1(*)] [10].

ATi_2O_5 undergoes an antiferromagnetic (AF) transition at $T_N \sim 26$ K for $CoTi_2O_5$ and $T_N \sim 40$ K for $FeTi_2O_5$ [8–11]. For $CoTi_2O_5$, the long-range AF order with the propagation vector $\mathbf{q} = (\pm\frac{1}{2}, \frac{1}{2}, 0)$ has been identified in neutron powder diffraction and muon-spin rotation experiments [8]. The emergence of this AF order requires a symmetry-lowering lattice distortion for the orthorhombic $CoTi_2O_5$. Thus, this orbital-nondegenerate frustrated antiferromagnet is suggested to be a spin-JT magnet. For $FeTi_2O_5$, by combining muon spin rotation and x-ray diffraction experiments with density functional theory calculations, it has been demonstrated that the crystal and AF structures are the same as those of $CoTi_2O_5$ [9]. This suggests that not only $CoTi_2O_5$ but also $FeTi_2O_5$ are spin-JT magnets. Although the symmetry-lowering lattice distortion in the AF state has not yet been experimentally resolved in either $CoTi_2O_5$ or $FeTi_2O_5$, recent resonant elastic x-ray scattering experiments in $CoTi_2O_5$ and thermal expansion and magnetostriction measurements in $FeTi_2O_5$ revealed the presence of magnetoelastic coupling, which supports the spin-JT scenario in ATi_2O_5 [10,12].

In this paper, we present ultrasound velocity measurements of the low-symmetry orthorhombic ATi_2O_5 ($A = Co, Fe$), from which we determine the elastic moduli of these

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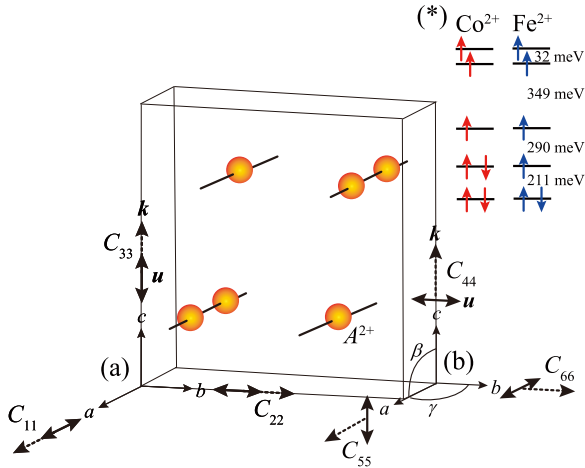


FIG. 1. Magnetic A^{2+} sites in ATi_2O_5 ($A = \text{Co, Fe}$) within the orthorhombic crystal unit cell, forming 1D a -axis chains (the solid lines). (*) Schematic energy levels of $\text{Co}^{2+} 3d^7$ and $\text{Fe}^{2+} 3d^6$ electrons with C_{2v} site symmetry in ATi_2O_5 . The values in (*) represent the crystal-field splitting energies in FeTi_2O_5 as reported in Ref. [10]. With the crystal axes (a) and (b), the propagation vector \mathbf{k} and polarization vector \mathbf{u} of the sound waves for (a) compressive elastic moduli C_{11} , C_{22} , and C_{33} and (b) shear elastic moduli C_{44} , C_{55} , and C_{66} are indicated. The angles β and γ between the crystal axes (b) correspond to the shear angles that are respectively tilted by the sound waves for C_{55} ($\mathbf{k} \parallel \mathbf{a}, \mathbf{u} \parallel \mathbf{c}$) and C_{66} ($\mathbf{k} \parallel \mathbf{b}, \mathbf{u} \parallel \mathbf{a}$).

compounds. The sound velocity or elastic modulus is a useful probe with which to extract symmetry-resolved thermodynamic information from a crystal [13]. Furthermore, as the ultrasound velocity can be measured with a high precision of approximately parts per million, its measurements can sensitively probe elastic anomalies driven by phase transitions, fluctuations, and excitations [13]. In magnets, the modified sound dispersions caused by magnetoelastic coupling allow the extraction of detailed information on the interplay of the lattice, spin, and orbital degrees of freedom [13–32].

In the spin-JT system, spin-lattice coupling induces a structural transition that lowers the crystal symmetry. For instance, the representative spin-JT system of chromite spinel ACr_2O_4 ($A = \text{Mg, Zn}$) has an AF transition at $T_N \sim 13$ K coinciding with the cubic-to-tetragonal lattice distortion [5,6]. For ACr_2O_4 , the ultrasound velocity measurements in the single crystal revealed that the temperature (T) dependence of the tetragonal shear modulus $(C_{11} - C_{12})/2$ exhibits Curie-type ($\sim -1/T$ -type) softening on cooling in the cubic paramagnetic phase ($T > T_N$), which is a precursor to the cubic-to-tetragonal lattice distortion at T_N [14]. This elastic anomaly above T_N indicates the presence of spin-lattice-coupled fluctuations in the paramagnetic phase of ACr_2O_4 , which is referred to as the dynamical spin-JT effect. In the present study of the orthorhombic pseudobrookites CoTi_2O_5 and FeTi_2O_5 , we find the presence of unusual elastic softness in CoTi_2O_5 in the paramagnetic phase above T_N , suggesting the emergence of dynamical spin-lattice-coupled state.

II. EXPERIMENTAL

Single crystals of CoTi_2O_5 with $T_N \sim 26$ K and FeTi_2O_5 with $T_N \sim 40$ K were grown adopting the floating-zone

TABLE I. Elastic moduli for ATi_2O_5 ($A = \text{Co, Fe}$) with an orthorhombic crystal structure, and the corresponding sound mode (propagation vector \mathbf{k} and polarization vector \mathbf{u}) and irreducible representation (irrep).

Elastic modulus	Sound mode (\mathbf{k} and \mathbf{u})	Irrep
C_{11}	Longitudinal wave ($\mathbf{k} \parallel \mathbf{u} \parallel \mathbf{a}$)	A_g
C_{22}	Longitudinal wave ($\mathbf{k} \parallel \mathbf{u} \parallel \mathbf{b}$)	A_g
C_{33}	Longitudinal wave ($\mathbf{k} \parallel \mathbf{u} \parallel \mathbf{c}$)	A_g
C_{44}	Transverse wave ($\mathbf{k} \parallel \mathbf{c}, \mathbf{u} \parallel \mathbf{b}$)	B_{3g}
C_{55}	Transverse wave ($\mathbf{k} \parallel \mathbf{a}, \mathbf{u} \parallel \mathbf{c}$)	B_{2g}
C_{66}	Transverse wave ($\mathbf{k} \parallel \mathbf{b}, \mathbf{u} \parallel \mathbf{a}$)	B_{1g}

method [8,9]. The ultrasound velocities were measured adopting the phase-comparison technique with longitudinal and transverse sound waves at a frequency of 30 MHz, where the ultrasound velocity or elastic modulus can be measured with a high precision of approximately parts per million. The ultrasound waves were generated and detected by LiNbO_3 transducers with a fundamental frequency of $f = 30$ MHz, which were attached to parallel mirror surfaces of the crystal, oriented perpendicular to the orthorhombic a , b , and c axes. The variation in ultrasound attenuation was monitored by observing the amplitude of the first transmitted echo. In the present experiments, ultrasound signals at higher-harmonic frequencies were not detected. Measurements were taken to determine the symmetrically independent elastic moduli of the orthorhombic crystal, specifically the compressive elastic moduli C_{11} , C_{22} , and C_{33} , and the shear elastic moduli C_{44} , C_{55} , and C_{66} (see Table I). In Fig. 1, the propagation vector \mathbf{k} and polarization vector \mathbf{u} of the sound waves for the respective elastic moduli are indicated along with the magnetic A^{2+} sites of ATi_2O_5 ($A = \text{Co, Fe}$) in the orthorhombic crystal unit cell, which form the 1D a -axis chains. As indicated in Fig. 1(a), the longitudinal sound wave corresponding to the compressive elastic modulus C_{11} propagates along the magnetic A^{2+} chains ($\mathbf{k} \parallel \mathbf{a}$), whereas the longitudinal waves corresponding to the compressive moduli C_{22} and C_{33} propagate orthogonal to the A^{2+} chains ($\mathbf{k} \perp \mathbf{a}$). Likewise, in Fig. 1(b), the transverse sound wave corresponding to the shear elastic modulus C_{55} propagates along the A^{2+} chains ($\mathbf{k} \parallel \mathbf{a}$), whereas the transverse waves corresponding to the shear C_{44} and C_{66} propagate orthogonal to the A^{2+} chains ($\mathbf{k} \perp \mathbf{a}$). The sound velocities of CoTi_2O_5 (FeTi_2O_5) measured at a room temperature of 300 K are 8820 m/s (8600 m/s) for C_{11} , 8750 m/s (6380 m/s) for C_{22} , 8630 m/s (7620 m/s) for C_{33} , 3920 m/s (3600 m/s) for C_{44} , 4300 m/s (4090 m/s) for C_{55} , and 4040 m/s (3660 m/s) for C_{66} .

III. RESULTS

A. CoTi_2O_5

Figures 2(a)–2(c) respectively present the temperature (T) dependence of the compressive elastic moduli $C_{11}(T)$, $C_{22}(T)$, and $C_{33}(T)$ in CoTi_2O_5 . $C_{11}(T)$ [Fig. 2(a)] and $C_{33}(T)$ [Fig. 2(c)] exhibit monotonic hardening on cooling from 300 to 2 K, as is usually observed in solids [33]. In addition, $C_{22}(T)$ [Fig. 2(b)] exhibits ordinary monotonic hardening on

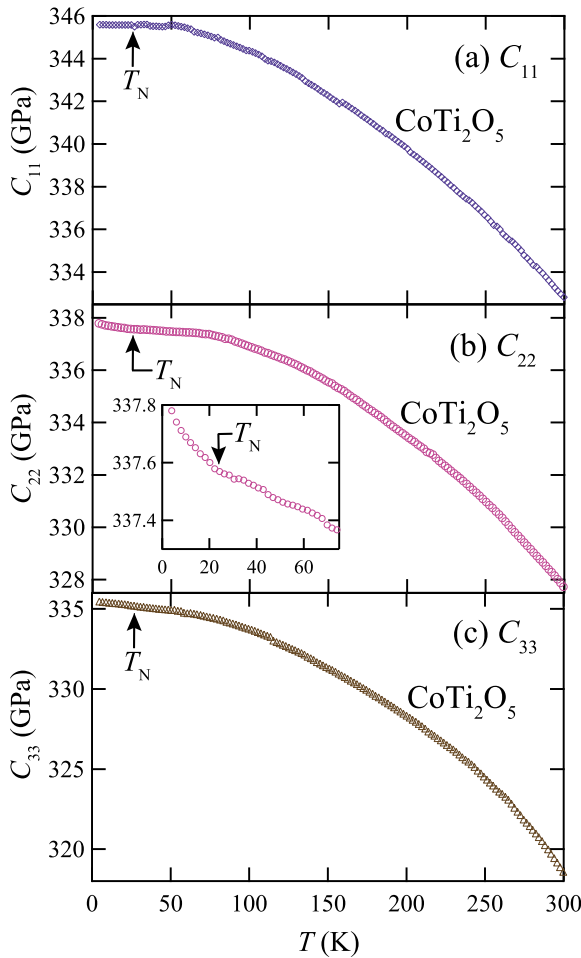


FIG. 2. Compressive elastic moduli of CoTi_2O_5 as functions of T : (a) $C_{11}(T)$, (b) $C_{22}(T)$, and (c) $C_{33}(T)$. The inset in (b) shows the expanded view of $C_{22}(T)$ below 75 K. The labeled arrows indicate $T_N \sim 26$ K of CoTi_2O_5 .

cooling from 300 to $T_N \sim 26$ K and a small slope change of hardening at $T_N \sim 26$ K.

Figures 3(a)–3(c) respectively depict the T dependence of the shear elastic moduli $C_{44}(T)$, $C_{55}(T)$, and $C_{66}(T)$ in CoTi_2O_5 . In these plots, all the elastic moduli exhibit hardening on cooling from 300 to ~ 60 K, but below ~ 60 K, only $C_{55}(T)$ [Fig. 3(b)] exhibits Curie-type ($\sim 1/T$ -type) softening on cooling to $T_N \sim 26$ K. Furthermore, $C_{55}(T)$ [Fig. 3(b)] and $C_{66}(T)$ [Fig. 3(c)] exhibit a discontinuous anomaly at $T_N \sim 26$ K, whereas there is no anomaly in $C_{44}(T)$ [Fig. 3(a)]. Near and below T_N , the ultrasound signals for $C_{55}(T)$ and $C_{66}(T)$ are attenuated but remain strong enough to accurately measure the T dependence of the absolute values of the sound velocities. As highlighted in the inset of Fig. 3(c), the T dependence of the ultrasound attenuation for C_{66} exhibits a sharp peak at $T_N \sim 26$ K, coinciding with the dip anomaly observed in $C_{66}(T)$.

In $C_{55}(T)$ [Fig. 3(b)], the Curie-type softening ceases below T_N , but the elasticity does not recover in the AF state below T_N , where the detected ultrasound signal is attenuated. In the magnetically ordered state, magnetostriction can induce domain-wall stress, resulting in a loss of elasticity [34]. Thus,

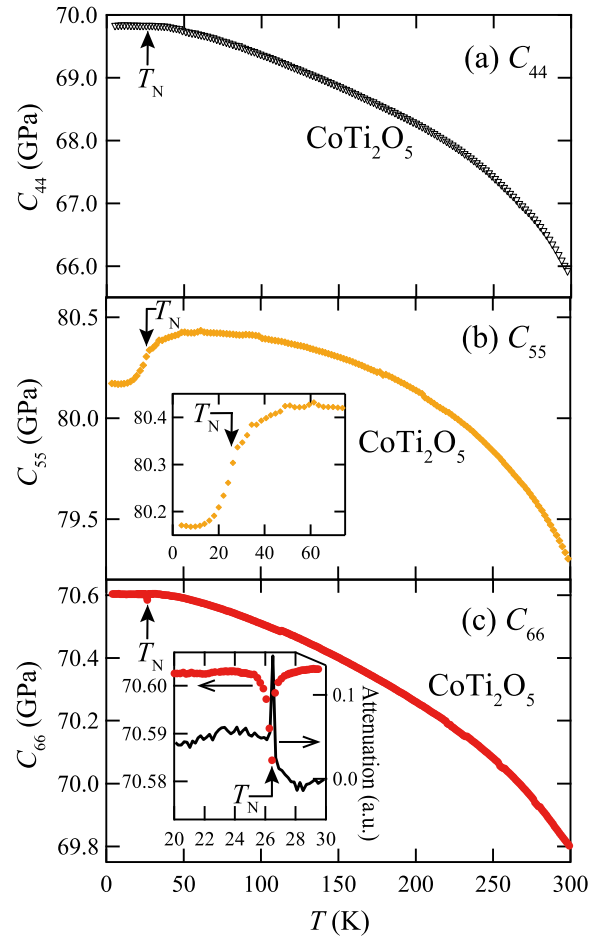


FIG. 3. Shear elastic moduli of CoTi_2O_5 as functions of T : (a) $C_{44}(T)$, (b) $C_{55}(T)$, and (c) $C_{66}(T)$. The inset in (b) is the expanded view of $C_{55}(T)$ below 75 K. The inset in (c) displays the expanded view of $C_{66}(T)$ (markers) along with the relative T dependence of the ultrasound attenuation for C_{66} normalized to its value at 30 K (solid line), in the range $20 \text{ K} \leq T \leq 30 \text{ K}$. The labeled arrows indicate $T_N \sim 26$ K of CoTi_2O_5 .

the softened elasticity observed in $C_{55}(T)$ below T_N can be attributed to stress effects on the magnetic domain walls.

B. FeTi_2O_5

Figures 4(a)–4(c) present the T dependence of the compressive elastic moduli $C_{11}(T)$, $C_{22}(T)$, and $C_{33}(T)$, respectively, in FeTi_2O_5 . $C_{11}(T)$ [Fig. 4(a)] and $C_{22}(T)$ [Fig. 4(b)] exhibit monotonic hardening on cooling from 300 to 2 K, as is usually observed in solids [33]. Moreover, $C_{33}(T)$ [Fig. 4(c)] exhibits ordinary monotonic hardening on cooling from 300 to $T_N \sim 40$ K and a small slope change of the hardening at $T_N \sim 40$ K.

Figures 5(a)–5(c) depict the T dependence of the shear elastic moduli $C_{44}(T)$, $C_{55}(T)$, and $C_{66}(T)$, respectively, in FeTi_2O_5 . $C_{66}(T)$ [Fig. 5(c)] exhibits monotonic hardening on cooling from 300 to 2 K. Moreover, $C_{44}(T)$ [Fig. 5(a)] and $C_{55}(T)$ [Fig. 5(b)] exhibit monotonic hardening on cooling from 300 K to $T_N \sim 40$ K and a small slope change of the hardening at $T_N \sim 40$ K.

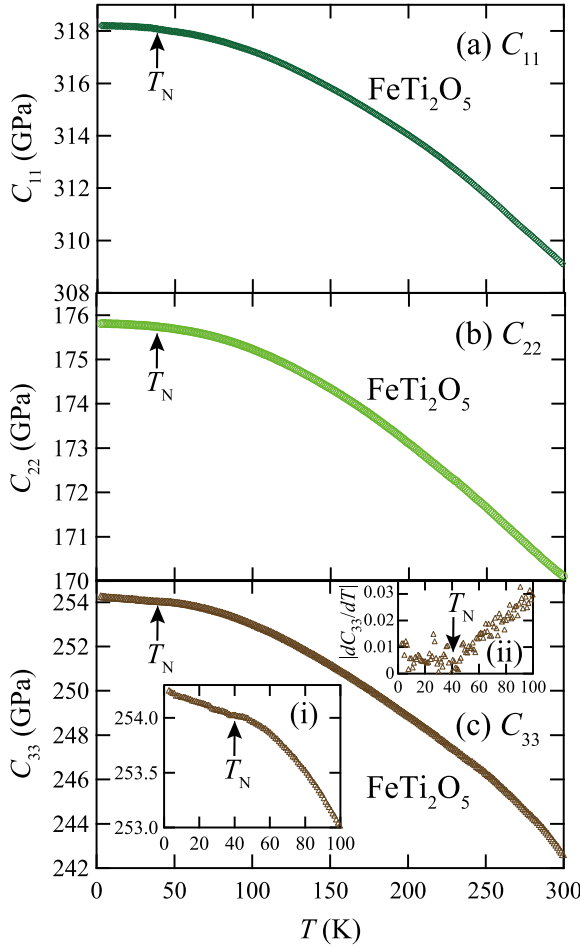


FIG. 4. Compressive elastic moduli of FeTi_2O_5 as functions of T : (a) $C_{11}(T)$, (b) $C_{22}(T)$, and (c) $C_{33}(T)$. The insets (i) and (ii) in (c) respectively show the expanded views of $C_{33}(T)$ and $|dC_{33}(T)/dT|$ below 100 K. The labeled arrows indicate $T_N \sim 40$ K of FeTi_2O_5 .

IV. DISCUSSION

A. Elastic anomalies in $C_{\Gamma}(T)$

For ATi_2O_5 ($A = \text{Co}, \text{Fe}$), ultrasound velocity measurements revealed the Curie-type softening in $C_{55}(T)$ of CoTi_2O_5 above T_N , which ceases below T_N [Fig. 3(b)], as well as the discontinuous anomalies at T_N in $C_{\Gamma}(T)$ for ATi_2O_5 [Figs. 2–5]. These elastic anomalies, observed above and at T_N , are likely of magnetic origin rather than being caused by oxygen cluster multipoles. Given the absence of orbital degeneracy at the A^{2+} site [Fig. 1(*), the spin degrees of freedom play a crucial role in these elastic behaviors. Such anomalies are attributed to magnetoelastic coupling acting on the exchange interactions, where the exchange striction arises from a modulation of the exchange interactions by ultrasound as follows [13]:

$$H_{\text{exs}} = \sum_{ij} [J(\delta + \mathbf{u}_i - \mathbf{u}_j) - J(\delta)] \mathbf{S}_i \cdot \mathbf{S}_j. \quad (1)$$

Here $\delta = \mathbf{R}_i - \mathbf{R}_j$ is the distance between two magnetic ions and \mathbf{u}_i is the displacement vector for the ion with equilibrium position \mathbf{R}_i . When a sound wave with polarization \mathbf{u} and

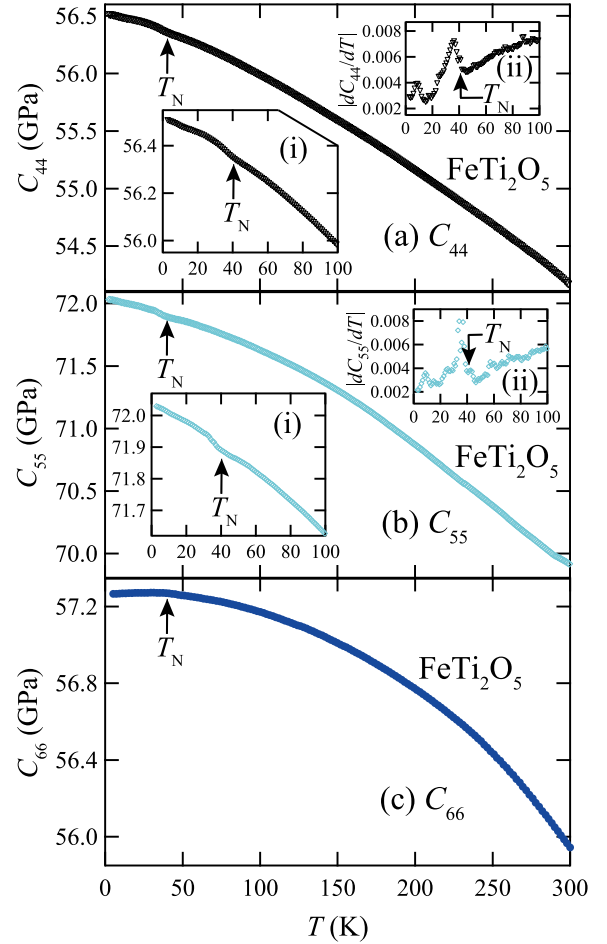


FIG. 5. Shear elastic moduli of FeTi_2O_5 as functions of T : (a) $C_{44}(T)$, (b) $C_{55}(T)$, and (c) $C_{66}(T)$. The insets (i) and (ii) in (a) and (b) respectively show the expanded views of $C_{55}(T)$, $|dC_{55}(T)/dT|$, $C_{66}(T)$, and $|dC_{66}(T)/dT|$ below 100 K. The labeled arrows indicate $T_N \sim 40$ K of FeTi_2O_5 .

propagation \mathbf{k} is given by $\mathbf{u} = \mathbf{u}_0 \exp[i(\mathbf{k} \cdot \mathbf{r} - \omega t)]$, where \mathbf{u}_0 and ω are the respective amplitude and frequency, the exchange striction of Eq. (1) is rewritten as [35]

$$H_{\text{exs}} = \sum_i \left(\frac{\partial J}{\partial \delta} \cdot \mathbf{u}_0 \right) (\mathbf{k} \cdot \delta) (\mathbf{S}_i \cdot \mathbf{S}_{i+\delta}) e^{i(\mathbf{k} \cdot \mathbf{R}_i - \omega t)}. \quad (2)$$

Here the exponential is expanded to first order because with a 30-MHz ultrasound frequency $k\delta \ll 1$. Equation (2) indicates that both the longitudinal and transverse sound waves can couple to the spin system via the exchange striction mechanism, depending on the directions of polarization \mathbf{u} and propagation \mathbf{k} relative to the exchange path δ .

For ATi_2O_5 with 1D A^{2+} chains, one possible origin for the observed elastic anomalies is the spin-Peierls effect, where the lattice distortion along the spin chain direction occurs at the transition temperature T_{sp} . In the T dependence of the elastic moduli of the spin-Peierls system, it is expected from the exchange striction mechanism [Eq. (2)] that only the compressive modulus along the spin chain direction has a pronounced anomaly at T_{sp} , and the shear moduli have no anomaly, as observed for the prototypical spin-Peierls compound CuGeO_3

[30–32]. Thus, for ATi_2O_5 , although a quasi-1D magnetic character has been experimentally suggested [11], the absence of an elastic anomaly at T_N in the spin-chain-direction (a -axis) compressive modulus $C_{11}(T)$ [Figs. 2(a) and 4(a)] rules out a possible spin-Peierls transition at T_N . In addition, the Curie-type softening in the shear modulus $C_{55}(T)$ of CoTi_2O_5 above T_N [Fig. 3(b)] should have an origin other than the spin-Peierls instability.

B. Curie-type softening in $C_\Gamma(T)$

In magnets, Curie-type softening in the T dependence of the elastic modulus $C_\Gamma(T)$ emerges as a precursor to a structural transition, which is driven by the coupling of the lattice to the electronic degrees of freedom [13,14,18,19,22–29]. Therefore, the Curie-type softening observed in the symmetry-lowering elastic mode $C_{55}(T)$ of CoTi_2O_5 above T_N should be a precursor to the symmetry-lowering lattice distortion at T_N . Such a precursor softening to the structural transition is well known to occur as a result of the Jahn-Teller effect in orbital-degenerate systems [13]. However, the absence of orbital degeneracy at the Co^{2+} site in CoTi_2O_5 rules out such orbital effects [Fig. 1(*)]. Thus, the Curie-type softening in $C_{55}(T)$ of CoTi_2O_5 above T_N is most likely driven by spin-lattice-coupled fluctuations, which are a precursor to a spin-driven magnetostructural transition at T_N . It is noted that the magnitude of the Curie-type softening in CoTi_2O_5 ($\Delta C_{55}/C_{55} \sim 0.15\%$) [Fig. 3(b)] is significantly smaller than that observed in the prototypical spin-JT system of chromite spinel ACr_2O_4 ($\Delta C_\Gamma/C_\Gamma \sim 50\%$) [14]. For CoTi_2O_5 , this relatively small softening above T_N is consistent with the fact that the lattice distortion below T_N is too small to have been resolved experimentally [8].

For CoTi_2O_5 , considering that the strain generated by ultrasound in C_{55} ($\mathbf{k} \parallel \mathbf{a}$, $\mathbf{u} \parallel \mathbf{c}$) tilts the monoclinic angle β between the a and c crystal axes [Fig. 1(b)], the Curie-type softening in $C_{55}(T)$ above T_N should be a precursor to the ac -plane shear lattice distortion at T_N . It is important to note that, for CoTi_2O_5 , the observed Curie-type softening in the ac -plane shear mode $C_{55}(T)$ above T_N is inconsistent with the structural symmetry breaking caused by AF ordering at T_N with propagation vector $\mathbf{q} = (\pm\frac{1}{2}, \frac{1}{2}, 0)$ [8]. For the magnetostructural transition to the AF order with $\mathbf{q} = (\pm\frac{1}{2}, \frac{1}{2}, 0)$, it is expected that an ab -plane shear lattice distortion occurs at T_N , where the monoclinic angle γ between the a and b crystal axes is tilted [Fig. 1(b)]. Therefore, for CoTi_2O_5 , the observation of the Curie-type softening in $C_{55}(T)$ above T_N suggests that the AF transition at T_N coincides with a orthorhombic-to-triclinic lattice distortion, which tilts the angles β and γ [Fig. 1(b)].

In CoTi_2O_5 , although the AF order with $\mathbf{q} = (\pm\frac{1}{2}, \frac{1}{2}, 0)$ is expected to coincide with the ab -plane shear lattice distortion at T_N , the ab -plane shear elastic modulus $C_{66}(T)$ shows no evidence of Curie-type softening above T_N [Fig. 3(c)]. In the spin-lattice-coupled system undergoing a magnetostructural transition, the T dependence of the elastic modulus $C_\Gamma(T)$ in the paramagnetic phase can be decomposed into a “usual” background component, $C_\Gamma^{(0)}(T)$, arising from lattice anharmonicity, and an “unusual” Curie component, $\delta C_\Gamma^C(T)$, originating from magnetostructural fluctuations:

$C_\Gamma(T) = C_\Gamma^{(0)}(T) + \delta C_\Gamma^C(T)$. Here $C_\Gamma^{(0)}(T)$ exhibits monotonic hardening with decreasing T [33], while $\delta C_\Gamma^C(T)$ contributes to the Curie-type softening, following $\delta C_\Gamma^C(T) \sim -1/T$ [14]. In the case of CoTi_2O_5 , the absence of observed Curie-type softening in $C_{66}(T)$ suggests that the contribution of the Curie component $\delta C_{66}^C(T)$, if present, is small and comparable to or smaller than that of the background $C_{66}^{(0)}(T)$.

For FeTi_2O_5 , the absence of observed Curie-type softening in $C_\Gamma(T)$ [Figs. 4 and 5] suggests that the contribution of the Curie component $\delta C_\Gamma^C(T)$ is either negligible or smaller than that in CoTi_2O_5 . However, previous experimental and theoretical studies have shown that its crystal and AF structures are identical to those of CoTi_2O_5 [9], and earlier experimental evidence has verified the presence of magnetoelastic coupling [10]. Thus, it is reasonable to expect a magnetostructural transition at T_N in FeTi_2O_5 , similar to that in CoTi_2O_5 , although the spin-lattice coupling in FeTi_2O_5 is likely weaker than in CoTi_2O_5 .

In the spin-JT system, the T dependence of the elastic modulus $C_\Gamma(T)$ in the paramagnetic phase is explained by assuming a coupling of ultrasound with the magnetic ions through the exchange striction mechanism [Eq. (2)] [13,14,22]. Specifically, the Curie component $\delta C_\Gamma^C(T)$ in $C_\Gamma(T)$ of the spin-JT system is explained by assuming the coupling of ultrasound to the structural unit cells via the exchange striction mechanism and the presence of exchange-striction-sensitive inter-unit-cell interactions. For the Curie-component-active elastic modulus $C_\Gamma(T) = C_\Gamma^{(0)}(T) + \delta C_\Gamma^C(T)$ in the prototypical spin-JT system ACr_2O_4 , the contribution of the Curie component $\delta C_\Gamma^C(T)$ is so large that the background component $C_\Gamma^{(0)}(T)$ can be assumed to be T independent: $C_\Gamma^{(0)}(T) \simeq C_\Gamma^{(0)}(0)$ with $C_\Gamma^{(0)}(0)$ the background component at $T = 0$ K [14]. In this case, the mean-field expression of $C_\Gamma(T)$ is written as

$$C_\Gamma(T) = C_\Gamma^{(0)}(0) \frac{T - T_c}{T - \theta}. \quad (3)$$

Here θ is the inter-unit-cell interaction, and $T_c = \theta + NG^2/C_\Gamma^{(0)}(0)$ is the second-order critical temperature for elastic softening $C_\Gamma \rightarrow 0$ with N the number density of the structural unit cell and G the constant of coupling of the structural order parameter to the strain. θ is positive (negative) when the interaction is ferrodistorptive (antiferrodistorptive). Equation (3) reproduces the observed Curie-type softening in $C_\Gamma(T)$ of ACr_2O_4 very well [14].

In Fig. 6, a fit of Eq. (3) to the experimental $C_{55}(T)$ of CoTi_2O_5 in the T range $T_N \leq T \leq 60$ K is shown as a solid curve, with the corresponding fit parameters listed. The fit curve aligns well with the experimental data within this T range. However, deviations from the fit become evident above ~ 60 K, indicating that the T -dependent background $C_{55}^{(0)}(T)$ contributes to the experimental $C_{55}(T)$ in CoTi_2O_5 .

For CoTi_2O_5 , the small observed softening in $C_{55}(T)$ ($\Delta C_{55}/C_{55} \sim 0.15\%$), combined with the deviation of the experimental $C_{55}(T)$ from the Eq. (3) fit above ~ 60 K (Fig. 6), suggests that the contribution of the Curie component $\delta C_{55}^C(T)$ is small and comparable to that of the background $C_{55}^{(0)}(T)$. Thus, for the Curie-component-active elastic modulus $C_\Gamma(T) = C_\Gamma^{(0)}(T) + \delta C_\Gamma^C(T)$ in ATi_2O_5 ($A = \text{Co}, \text{Fe}$), we

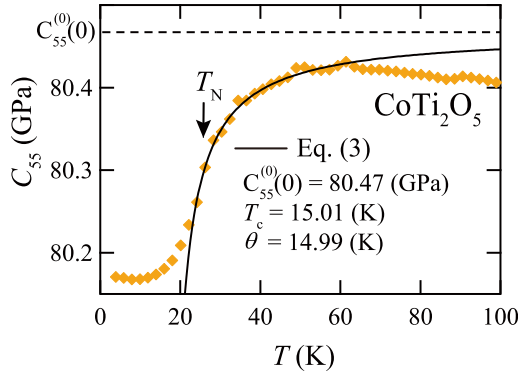


FIG. 6. $C_{55}(T)$ below 100 K in CoTi_2O_5 [markers from Fig. 3(b)]. The labeled arrows indicate $T_N \sim 26$ K of CoTi_2O_5 . The solid curve is a fit of Eq. (3) to the experimental $C_{55}(T)$ in the T range $T_N \leq T \leq 60$ K; the values for the fit parameters are also listed. The dashed horizontal line indicates $C_{55}^{(0)}(0)$ in Eq. (3).

should consider the T -dependent background $C_{\Gamma}^{(0)}(T)$. In this case, Eq. (3) is rewritten as

$$C_{\Gamma}(T) = C_{\Gamma}^{(0)}(T) - \frac{NG^2}{T - \theta}, \quad (4)$$

where $\delta C_{\Gamma}^C(T) = -\frac{NG^2}{T - \theta}$. For the experimental $C_{\Gamma}(T)$ in ATi_2O_5 , it is unfortunately not possible in the present study to determine the “true” T -dependent background $C_{\Gamma}^{(0)}(T)$. Consequently, the experimental $C_{\Gamma}(T)$ cannot be analyzed using Eq. (4). In future work, measurements of $C_{\Gamma}(T)$ in the nonmagnetic analog of the orthorhombic pseudobrookite MgTi_2O_5 might help determine the background $C_{\Gamma}^{(0)}(T)$ for $C_{\Gamma}(T)$ in ATi_2O_5 [36], enabling a more accurate analysis of the experimental $C_{\Gamma}(T)$ for ATi_2O_5 using Eq. (4).

C. Non-Curie-type $C_{\Gamma}(T)$

For ATi_2O_5 ($A = \text{Co, Fe}$), this study finds that only $C_{55}(T)$ of CoTi_2O_5 exhibits Curie-type softening above T_N , while other elastic moduli, $C_{\Gamma}(T)$, show hardening on cooling [Figs. 2–5]. However, as discussed in Sec. IV B, the small magnitude of the observed softening in $C_{55}(T)$ of CoTi_2O_5 ($\Delta C_{55}/C_{55} \sim 0.15\%$) suggests that the contribution of the Curie component $\delta C_{55}^C(T)$ is small and comparable to that of the background $C_{55}^{(0)}(T)$. Therefore, it is necessary to investigate the potential presence of the Curie component $\delta C_{\Gamma}^C(T)$ in the “non-Curie-type” elastic moduli $C_{\Gamma}(T)$ of ATi_2O_5 , excluding the Curie-type $C_{55}(T)$ of CoTi_2O_5 .

For the non-Curie-type elastic moduli $C_{\Gamma}(T)$ of ATi_2O_5 , we investigate the potential presence of the Curie component $\delta C_{\Gamma}^C(T)$ by fitting the experimental $C_{\Gamma}(T)$ to an empirical equation for the background $C_{\Gamma}^{(0)}(T)$ [33]:

$$C_{\Gamma}^{(0)}(T) = C_{\Gamma}^{(0)}(0) - \frac{A}{\exp(B/T) - 1}, \quad (5)$$

where $C_{\Gamma}^{(0)}(0)$ is the background elastic modulus at $T = 0$ K and A and B are fitting parameters. In the case of a Curie-component-active elastic modulus, $C_{\Gamma}(T) = C_{\Gamma}^{(0)}(T) + \delta C_{\Gamma}^C(T)$, the contribution of the Curie component, $\delta C_{\Gamma}^C(T) \sim -1/T$, to $C_{\Gamma}(T)$ weakens at higher T 's, but strengthens

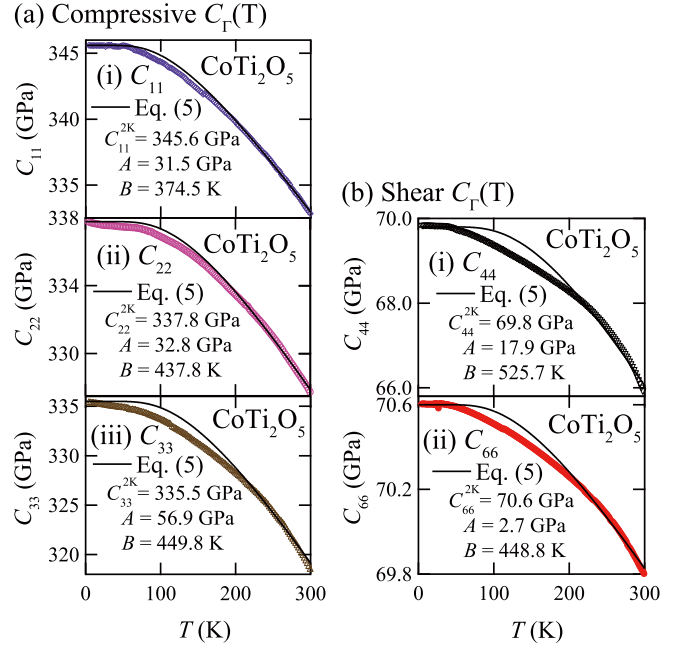


FIG. 7. $C_{\Gamma}(T)$ of CoTi_2O_5 : (a) Compressive $C_{\Gamma}(T)$ of (i) $C_{11}(T)$, (ii) $C_{22}(T)$, and (iii) $C_{33}(T)$ (markers, from Fig. 2) and (b) shear $C_{\Gamma}(T)$ of (i) $C_{44}(T)$ and (ii) $C_{66}(T)$ (markers, from Fig. 3). The solid curves are fits of Eq. (5) to the experimental $C_{\Gamma}(T)$ in the T range $200 \text{ K} \leq T \leq 300 \text{ K}$ with the assumption of $C_{\Gamma}^{(0)}(0) = C_{\Gamma}^{2K}$; the values for the fit parameters are also listed.

at lower T 's. Consequently, fitting Eq. (5) to the Curie-component-active experimental $C_{\Gamma}(T)$ should yield good agreement at higher T 's, but deviations at lower T 's where $C_{\Gamma}^{(0)}(T) > C_{\Gamma}(T)$. Therefore, we fit Eq. (5) to the non-Curie-type experimental $C_{\Gamma}(T)$ of ATi_2O_5 [Figs. 2–5, excluding Fig. 3(b)] in the higher T range $200 \text{ K} \leq T \leq 300 \text{ K}$, assuming that $C_{\Gamma}^{(0)}(0)$ in Eq. (5) equals the value of the experimental $C_{\Gamma}(T)$ at the lowest measured temperature (2 K), C_{Γ}^{2K} , i.e., $C_{\Gamma}^{(0)}(0) = C_{\Gamma}^{2K}$.

In Fig. 7, the fits of Eq. (5) to the non-Curie-type experimental $C_{\Gamma}(T)$ of CoTi_2O_5 , $C_{\Gamma}(T)$ excluding $C_{55}(T)$, in the T range $200 \text{ K} \leq T \leq 300 \text{ K}$ are shown as solid curves. The fitting parameter values are also listed in Fig. 7, where $C_{\Gamma}^{(0)}(0)$ in Eq. (5) is fixed at $C_{\Gamma}^{(0)}(0) = C_{\Gamma}^{2K}$. In Fig. 7, the fit curves for $C_{11}(T)$ [Fig. 7(a)(i)] and $C_{22}(T)$ [Fig. 7(a)(ii)] agree well with the experimental $C_{\Gamma}(T)$. However, the fit curves for $C_{33}(T)$ [Fig. 7(a)(iii)], $C_{44}(T)$ [Fig. 7(b)(i)], and $C_{66}(T)$ [Fig. 7(b)(ii)] begin to deviate from the experimental $C_{\Gamma}(T)$ below ~ 200 K. These deviations are likely attributed to the presence of the Curie component $\delta C_{\Gamma}^C(T)$ or a more complex background $C_{\Gamma}^{(0)}(T)$ that is not fully captured by Eq. (5), possibly due to oxygen cluster multipoles.

In Fig. 8, the solid curves represent fits of Eq. (5) to the non-Curie-type experimental $C_{\Gamma}(T)$ of FeTi_2O_5 in the T range $200 \text{ K} \leq T \leq 300 \text{ K}$. The fitting parameter values are also listed in Fig. 8, where $C_{\Gamma}^{(0)}(0)$ in Eq. (5) is fixed at $C_{\Gamma}^{(0)}(0) = C_{\Gamma}^{2K}$. The fit curves for $C_{11}(T)$ [Fig. 8(a)(i)] and $C_{22}(T)$ [Fig. 8(a)(ii)] agree well with the experimental $C_{\Gamma}(T)$, similar to the fits for $C_{11}(T)$ and $C_{22}(T)$ of CoTi_2O_5 [Figs. 7(a)(i) and 7(a)(ii), respectively]. Additionally, for

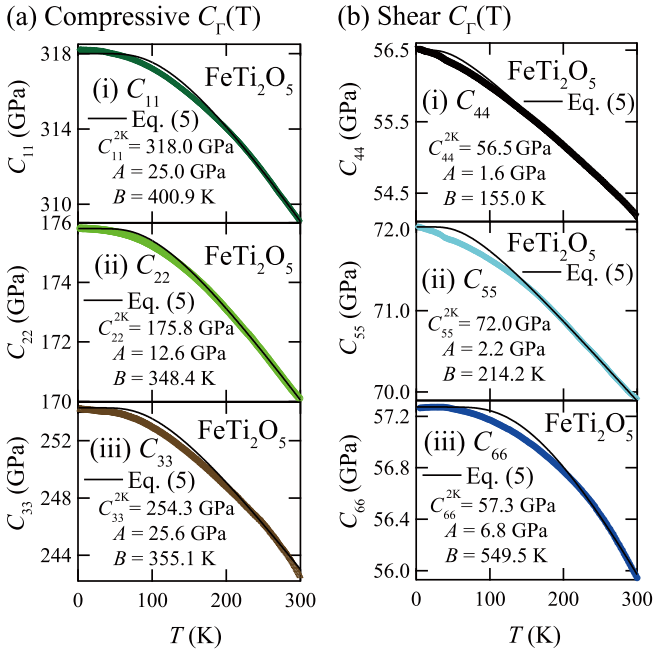


FIG. 8. $C_T(T)$ of FeTi_2O_5 : (a) Compressive $C_T(T)$ of (i) $C_{11}(T)$, (ii) $C_{22}(T)$, and (iii) $C_{33}(T)$ (markers, from Fig. 4) and (b) shear $C_T(T)$ of (i) $C_{44}(T)$, (ii) $C_{55}(T)$, and (iii) $C_{66}(T)$ (markers, from Fig. 5). The solid curves are fits of Eq. (5) to the experimental $C_T(T)$ in the T range $200 \text{ K} \leq T \leq 300 \text{ K}$ with the assumption of $C_T^{(0)}(0) = C_T^{2K}$; the values for the fit parameters are also listed.

FeTi_2O_5 , the deviations in the fits for $C_{33}(T)$ [Fig. 8(a)(iii)] and $C_{44}(T)$ [Fig. 8(b)(i)] are notably smaller than those for CoTi_2O_5 [Figs. 7(a)(iii) and 7(b)(i)]. Among the elastic moduli $C_{11}(T)$ – $C_{66}(T)$ of FeTi_2O_5 (Fig. 8), the fits for $C_{55}(T)$ [Fig. 8(b)(ii)] and $C_{66}(T)$ [Fig. 8(b)(iii)] exhibit noticeable deviations. These deviations are likely due to the presence of the Curie component $\delta C_T^C(T)$ or a more complex background $C_T^{(0)}(T)$ that is not fully described by Eq. (5), potentially caused by oxygen cluster multipoles. To address these deviations in FeTi_2O_5 , as well as the previously noted deviations in $C_{33}(T)$ [Fig. 7(a)(iii)], $C_{44}(T)$ [Fig. 7(b)(i)], and $C_{66}(T)$ [Fig. 7(b)(ii)] of CoTi_2O_5 , further studies are required. Future work should investigate whether these deviations originate from a dynamic elastic response or other factors, through analyses of the ultrasound frequency dependence of $C_T(T)$ and ultrasound attenuation.

D. Magnetostructural fluctuations

The present study reveals the presence of the Curie component, $\delta C_T^C(T)$, in $C_{55}(T)$ of CoTi_2O_5 , where the Curie-type softening is directly observed [Fig. 3(b)]. Additionally, as discussed in Sec. IV C, the fit of Eq. (5) to the non-Curie-type experimental $C_T(T)$ shows noticeable deviations in $C_{33}(T)$, $C_{44}(T)$, and $C_{66}(T)$ for CoTi_2O_5 (Fig. 7) and in $C_{55}(T)$ and $C_{66}(T)$ for FeTi_2O_5 (Fig. 8). This suggests the presence of either the Curie component $\delta C_T^C(T)$ or a more complex background $C_T^{(0)}(T)$ that Eq. (5) cannot fully capture.

For CoTi_2O_5 , as discussed in Sec. IV B, the observed Curie-type softening in the ac -plane shear modulus $C_{55}(T)$

above T_N [Fig. 3(b)] is inconsistent with the structural symmetry breaking caused by AF ordering at T_N with $\mathbf{q} = (\pm\frac{1}{2}, \frac{1}{2}, 0)$ [8]. This suggests that the lattice distortion at T_N involves not only the ac -plane shear but also the ab -plane shear, indicating the orthorhombic-to-triclinic lattice distortion at T_N . Therefore, it is expected for CoTi_2O_5 that the Curie-component $\delta C_T^C(T)$ is present in not only the ac -plane shear $C_{55}(T)$ but also the ab -plane shear $C_{66}(T)$, indicating the presence of two distinct types of magnetostructural fluctuations in CoTi_2O_5 above T_N : the ab -plane shear fluctuations and the ac -plane shear fluctuations. The ab -plane fluctuations are likely a precursor to the AF order with $\mathbf{q} = (\pm\frac{1}{2}, \frac{1}{2}, 0)$, consistent with the spin-JT scenario [8]. In contrast, the ac -plane fluctuations are newly identified in this study. Further experimental and theoretical investigations are needed to clarify the coexistence mechanism of the ab -plane and ac -plane shear magnetostructural fluctuations in CoTi_2O_5 .

In CoTi_2O_5 , according to the exchange striction mechanism described in Eq. (2), the transverse sound waves for $C_{55}(\mathbf{u} \perp \mathbf{a})$ and $C_{66}(\mathbf{k} \perp \mathbf{a})$ (Fig. 1) do not couple to the exchange interactions along the a axis. Instead, they couple to the inter- a -axis-chain exchange interactions. Consequently, the Curie components in $C_{55}(T)$ and $C_{66}(T)$ of CoTi_2O_5 above T_N likely originate from the coupling between the lattice and the inter- a -axis-chain exchange interactions. This suggests that, in CoTi_2O_5 , while the exchange interactions are quasi-1D in nature, the magnetostructural transition at T_N is driven by 3D spin-lattice coupling involving the inter- a -axis-chain exchange interactions.

For FeTi_2O_5 , the present study does not confirm the presence of magnetostructural fluctuations above T_N . However, previous experimental and theoretical studies have shown that its crystal and AF structures are identical to those of CoTi_2O_5 [9], and earlier experimental evidence has verified the presence of magnetoelastic coupling [10]. Therefore, it is reasonable to expect magnetostructural fluctuations in FeTi_2O_5 above T_N , similar to those in CoTi_2O_5 , although the spin-lattice coupling in FeTi_2O_5 is likely weaker than in CoTi_2O_5 . The difference in the strength of spin-lattice coupling between CoTi_2O_5 and FeTi_2O_5 is likely related to the distinct spin states of Co^{2+} ($3d^7$, $S = 3/2$) and Fe^{2+} ($3d^6$, $S = 2$) ions, respectively.

In the present study, the ultrasound velocity measurements were performed in zero magnetic field. Recent magnetostriction measurements on FeTi_2O_5 revealed that the intrachain a -axis and interchain b -axis exhibit negative and positive magnetostrictions, respectively, indicating the presence of magnetoelastic coupling [10]. This finding suggests that, in ATi_2O_5 ($A = \text{Co, Fe}$), the magnetic field is likely to influence the spin-lattice-coupled frustration. Ultrasound velocity measurements on ATi_2O_5 under magnetic field are anticipated to yield valuable insights into the effects of magnetic field on the frustration physics of these compounds.

V. SUMMARY

Ultrasound velocity measurements of the orthorhombic pseudobrookites CoTi_2O_5 and FeTi_2O_5 revealed unusual elastic softness in CoTi_2O_5 above T_N in the symmetry-lowering

shear elastic mode, specifically in the *ac*-plane shear modulus C_{55} . For CoTi_2O_5 , the observed elastic softness in C_{55} above T_N is inconsistent with the structural symmetry breaking caused by AF ordering at T_N . This suggests the presence of two distinct types of magnetostructural fluctuations above T_N , likely acting as a precursor to the symmetry-lowering lattice distortion at T_N . In contrast, the measurements for FeTi_2O_5 show that such elastic softness is either negligible or smaller, indicating weaker spin-lattice coupling. These findings highlight CoTi_2O_5 and FeTi_2O_5 as unique spin-lattice-coupled frustrated systems with low crystal symmetry, where, while the exchange interactions are quasi-1D in nature,

the frustration is released by further lowering the crystal symmetry through 3D spin-lattice coupling, which is stronger in CoTi_2O_5 than in FeTi_2O_5 .

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