

## Anisotropic magnetoelastic coupling in the honeycomb magnet $\text{Na}_3\text{Co}_2\text{SbO}_6$

Prashanta K. Mukharjee, Sebastian Erdmann, Lichen Wang, Julian Kaiser, Anton Jesche, Pascal Puphal, Masahiko Isobe, Matthias Hepting, Bernhard Keimer, Philipp Gegenwart, Alexander A. Tsirlin

### Angaben zur Veröffentlichung / Publication details:

Mukharjee, Prashanta K., Sebastian Erdmann, Lichen Wang, Julian Kaiser, Anton Jesche, Pascal Puphal, Masahiko Isobe, et al. 2026. "Anisotropic magnetoelastic coupling in the honeycomb magnet  $\text{Na}_3\text{Co}_2\text{SbO}_6$ ." *Physical Review B* 113 (20): 205148. <https://doi.org/10.1103/tn1m-sjdh>.

### Nutzungsbedingungen / Terms of use:

licgercopyright

Dieses Dokument wird unter folgenden Bedingungen zur Verfügung gestellt: / This document is made available under these conditions:

**Deutsches Urheberrecht**

Weitere Informationen finden Sie unter: / For more information see:

<https://www.uni-augsburg.de/de/organisation/bibliothek/publizieren-zitieren-archivieren/publiz/>



# Anisotropic magnetoelastic coupling in the honeycomb magnet $\text{Na}_3\text{Co}_2\text{SbO}_6$

Prashanta K. Mukharjee<sup>1,\*</sup>, Sebastian Erdmann<sup>1</sup>, Lichen Wang<sup>2</sup>, Julian Kaiser<sup>1</sup>, Anton Jesche<sup>1</sup>, Pascal Puphal<sup>2</sup>, Masahiko Isobe<sup>2</sup>, Matthias Hepting<sup>2</sup>, Bernhard Keimer<sup>2</sup>, Philipp Gegenwart<sup>1,†</sup> and Alexander A. Tsirlin<sup>3,‡</sup>

<sup>1</sup>Experimental Physics VI, Center for Electronic Correlations and Magnetism, Institute of Physics, University of Augsburg, 86159 Augsburg, Germany

<sup>2</sup>Max-Planck-Institute for Solid State Research, Heisenbergstraße 1, 70569 Stuttgart, Germany

<sup>3</sup>Felix Bloch Institute for Solid-State Physics, University of Leipzig, 04103 Leipzig, Germany



(Received 5 March 2026; revised 28 April 2026; accepted 5 May 2026; published 26 May 2026)

We present magnetization and dilatometry measurements on the honeycomb cobaltate  $\text{Na}_3\text{Co}_2\text{SbO}_6$  and map out its detailed field-temperature phase diagram down to sub-Kelvin temperatures. Our data for in-plane magnetic fields show a strongly anisotropic  $c^*$ -axis lattice response, which is dominated by the variation of Co–O–Co bond angles according to *ab initio* calculations. At  $T = 0.4$  K, the magnetization  $M(B)$  exhibits steplike features that are also highly anisotropic. In the case of  $B \parallel b$ , a small hysteresis observed around the second field-induced magnetic transition ( $B_{c2}$ ) indicates its first-order character, whereas the apparent divergence of the magnetic Grüneisen parameter at  $B_{c2}$  is suppressed upon cooling and signals the absence of quantum critical behavior upon entering the field-polarized state. None of our thermodynamic measurements provide evidence for a field-induced quantum spin-liquid state near or above  $B_{c2}$ .

DOI: [10.1103/tn1m-sjdh](https://doi.org/10.1103/tn1m-sjdh)

## I. INTRODUCTION

The number of candidate quantum spin-liquid (QSL) materials has been rapidly increasing in recent years. One important theoretical framework in this context is the Kitaev honeycomb model, which hosts an exactly solvable QSL ground state [1]. The connection between the QSL state and the Kitaev model has been pivotal, driving intense research over the past decade. Honeycomb compounds based on  $4d/5d$  transition metals, such as  $\alpha\text{-RuCl}_3$  and  $(\text{Li,Na})_2\text{IrO}_3$ , are leading examples, extensively studied as possible Kitaev QSL candidates [2]. Although these materials often show long-range antiferromagnetic (AFM) order due to additional interactions and deviations from ideal crystal symmetry, techniques like hydrostatic pressure [3], doping [4], and applied magnetic fields have been explored to suppress this order and realize a Kitaev QSL state [5]. However, realizing Kitaev QSL in real materials is challenging because purely bond-directional Kitaev interactions are easily overwhelmed by unavoidable Heisenberg and off-diagonal terms, as well as interactions beyond nearest neighbors.

Recently,  $3d^7$   $\text{Co}^{2+}$  compounds, such as  $\text{Na}_3\text{Co}_2\text{SbO}_6$ ,  $\text{Na}_2\text{Co}_2\text{TeO}_6$ , and  $\text{BaCo}_2(\text{AsO}_4)_2$ , have been proposed as new Kitaev candidates [6,7]. Theoretically, these honeycomb cobaltates are expected to host dominant Kitaev interactions because  $\text{Co}^{2+}$  is a  $J_{\text{eff}} = \frac{1}{2}$  pseudospin ion, whereas the nearly  $90^\circ$  Co–O–Co bonding geometry suppresses isotropic Heisenberg exchange and enhances bond-directional Kitaev terms.

At the same time, they offer a practical advantage over  $4d/5d$  systems, as they can be synthesized as large, high-quality single crystals with minimal structural disorder. These features render cobaltates an especially promising platform in comparison with their  $4d/5d$  counterparts to study Kitaev-related physics. However, the non-Kitaev interactions are also significant or even dominant in cobaltates due to the subtle interplay of various hopping channels [8,9].

Phenomenologically, the honeycomb cobaltates share several similarities with  $\alpha\text{-RuCl}_3$  as a typical  $4d/5d$  Kitaev material. Both the cobaltates and  $\alpha\text{-RuCl}_3$  develop antiferromagnetic long-range order (LRO) in zero field, but this order is easily suppressed by applying magnetic field within the honeycomb plane, potentially leading to a field-induced Kitaev QSL. In the case of  $\alpha\text{-RuCl}_3$ , the character of the field-induced state above the critical field remains an active topic of debate [10]. Field-induced behavior of the honeycomb cobaltates is also under an active scrutiny [11–14]. These materials often reveal distinct field-induced phases in the vicinity of the saturation field [15–20], but magnetic order appears to persist within such phases [15,21–23]. An additional complexity arises from the multi- $q$  states reported in zero field in some of the cobaltates [24], in contrast to the simple zigzag order observed in  $\alpha\text{-RuCl}_3$ .

Here, we focus on  $\text{Na}_3\text{Co}_2\text{SbO}_6$  (NCSO) that crystallizes in the monoclinic space group  $C2/m$  wherein edge-sharing  $\text{CoO}_6$  octahedra form a honeycomb network in the  $ab$  plane [see Fig. 1(a)] [25]. Upon cooling in zero field, NCSO undergoes an AFM LRO below  $T_N \simeq 7$  K with a highly anisotropic field-induced response in the  $ab$  plane [25]. Magnetic ground state in zero field was long believed to be a simple zigzag phase [26–28], but recent studies revealed a more intricate double- $q$  magnetic structure [25,29] with residual spin

\*Contact author: [pkmukharjee92@gmail.com](mailto:pkmukharjee92@gmail.com)

†Contact author: [philipp.gegenwart@physik.uni-augsburg.de](mailto:philipp.gegenwart@physik.uni-augsburg.de)

‡Contact author: [altsirlin@gmail.com](mailto:altsirlin@gmail.com)

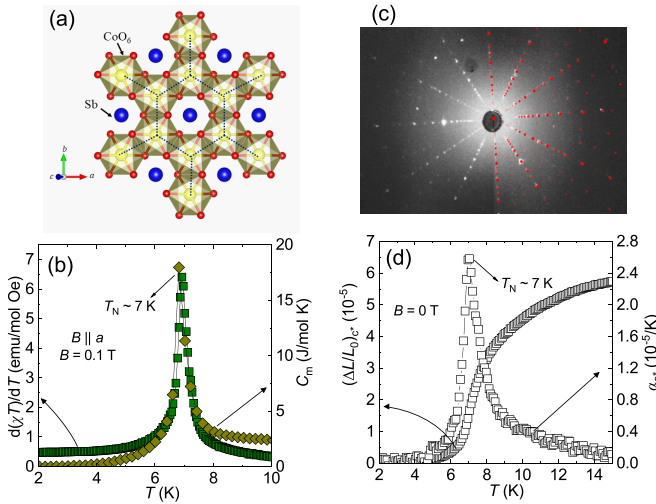


FIG. 1. (a) Crystal structure of  $\text{Na}_3\text{Co}_2\text{SbO}_6$ . The honeycomb network formed by  $\text{CoO}_6$  octahedra spread in the  $ab$  plane. (b) Fisher's heat capacity and magnetic specific heat vs  $T$  under zero magnetic field in the left and right  $y$  axes, respectively. (c) Left half: reflections from the Laue pattern taken on a twin-free crystal. Right half: simulation (red spots) of the Laue pattern using the CRYSTAL MAKER program. (d) Zero-field thermal expansion of NCSO.

dynamics [30]. The presence of such a multi- $q$  state, combined with pronounced in-plane anisotropy, renders NCSO a compelling system for studying the effect of applied field on a honeycomb magnet.

Several recent works have explored field-induced effects in NCSO. At  $T = 2$  K, in the ordered state, two distinct field-induced transitions are visible. The corresponding phase boundaries are anisotropic and appear at the critical fields  $B_{c1} \simeq 1.25$  T and  $B_{c2} \simeq 1.8$  T for  $B \parallel a$  vs  $B_{c1} \simeq 0.7$  T and  $B_{c2} \simeq 0.85$  T for  $B \parallel b$  orientations [25]. The region around  $B_{c2}$  is of particular interest and has been studied with different probes such as nuclear magnetic resonance (NMR) [31], THz spectroscopy [32], heat transport [33], ac magnetostriction [34], and magnetocaloric effect [35]. However, the coupling between spins and lattice in NCSO is still largely unexplored. Magnetoelastic probes, in particular thermal expansion and magnetostriction, are very sensitive to phase transitions and magnetic anisotropy and can reveal signatures of quantum critical behavior or even a field-induced QSL state [36].

These considerations raise three important questions for NCSO: (i) How strong is the magnetoelastic coupling, and how anisotropic is it with respect to the in-plane field direction? (ii) What is the nature of the field-induced transitions at  $B_{c1}$  and  $B_{c2}$ ? (iii) How do these transitions evolve toward  $T = 0$ ? Here, we address these questions by combining high-resolution dilatometry with other thermodynamic probes on high-quality twin-free single crystals of NCSO. We perform  $c^*$ -axis thermal expansion and magnetostriction measurements for magnetic fields applied along the in-plane  $a$  and  $b$  directions, and also report the magnetization and magnetic Grüneisen parameter in the sub-Kelvin temperature range. We quantify the magnetoelastic response across all the

field-induced changes and construct a detailed  $B$ - $T$  phase diagram. We find a pronounced in-plane anisotropy of the magnetoelastic coupling and clear signatures of the first-order behavior at  $B_{c1}$ . The apparent divergence of the magnetic Grüneisen parameter near  $B_{c2}$  is weak and does not increase at very low temperatures, thus giving evidence against a genuine field-induced quantum critical point in NCSO. Our results show that the intermediate-field regime of NCSO is governed by strongly anisotropic magnetic interactions and lacks any distinct signatures of a field-induced QSL state.

## II. METHODS

High-quality single crystals of NCSO were grown using an optimized flux method (see Supplemental Material, Ref. [37]). The quality of these crystals was rigorously verified through extensive Laue mapping (Fig. S1) and thermodynamic measurements. The NCSO crystals often exhibit twinned structural domains. For this study, we selected a twin-free crystal measuring  $1 \times 1 \times 0.1$  mm<sup>3</sup>. Laue analysis confirmed the absence of any structural domains in the chosen crystal. Notably, the twin-free crystal displayed only two field-induced features for each field direction (at 2 K), whereas twinned crystals exhibited multiple transitions (not shown) for a given direction of the applied field. Furthermore, angle-dependent susceptibility measurements confirm the absence of twinning in the investigated crystal, as evidenced by the clear twofold symmetry observed (see Fig. S8).

Magnetization measurements were performed using a Quantum Design Magnetic Property Measurement System (QD-MPMS3). For measurements conducted below 2 K, a <sup>3</sup>He attachment was utilized. Magnetic isotherms were recorded at a sweep rate of 200 Oe/sec, with the magnetic field stabilized at each field set point. Angle-dependent magnetization was recorded by employing the sample rotation stage of the MPMS3. The specific heat was measured using a standard relaxation technique within the QD Physical Property Measurement System (PPMS). The magnetic Grüneisen parameter was estimated from the magnetocaloric effect using an alternating-field technique [38].

Thermal expansion ( $\Delta L/L_0$ ) and magnetostriction ( $\lambda$ ) were measured along the  $c^*$  direction perpendicular to the  $ab$  plane using a high-resolution capacitance dilatometer following the procedure described in Ref. [39]. The temperature and field scans of the length change were carried out at rates of 0.3 K/min and 20 Oe/sec, respectively. Our dc measurements directly probe the out-of-plane expansion of NCSO, in contrast to Ref. [34] where in-plane lattice expansion was measured in the ac mode, resulting in the field derivative of the magnetostriction ( $d\lambda/dB$ ) and not the magnetostriction itself.

Effect of magnetic order on the lattice parameters and atomic structure of NCSO was studied by density-functional theory (DFT) band-structure calculations performed in the VASP code [40,41] using the Perdew-Burke-Ernzerhof version of the exchange-correlation potential [42]. All calculations included spin-orbit (SO) coupling and the mean-field correction for correlation effects in the Co 3d shell, with the onsite Coulomb repulsion  $U_d = 5$  eV and Hund's coupling  $J_d = 1$  eV [43].

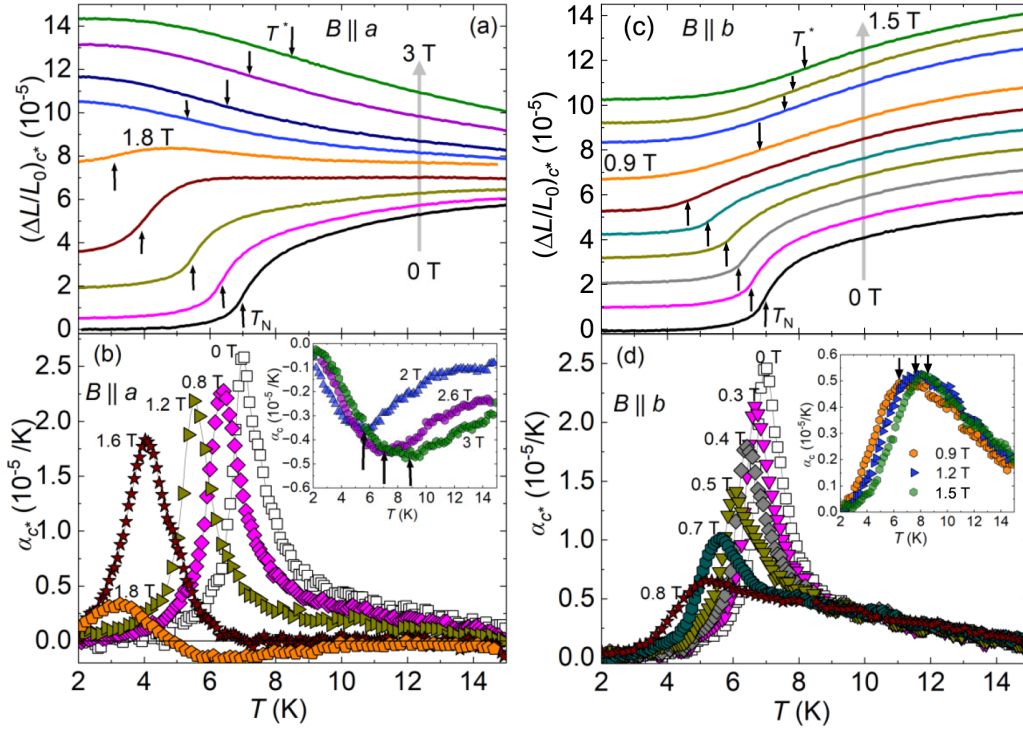


FIG. 2. (a), (c) Temperature dependence of the relative length changes for  $B \parallel a$  and  $B \parallel b$ , respectively (warming curves are shown). The upward arrows indicate the position of the  $T_N$ . The downward arrows mark the position of the crossover temperature  $T^*$  above  $B_{c2}$ . (b), (d) The corresponding plots of the linear TE coefficient,  $\alpha_{c^*}$  vs  $T$  for  $B \parallel a$  and  $B \parallel b$ , respectively. The insets show the data measured above  $B_{c2}$ . The arrows indicate the position of the broad maximum and minimum in  $\alpha_{c^*}$ .

### III. RESULTS

#### A. Thermal expansion

Figure 2 displays the thermal expansion (TE) data for NCSO up to 15 K, with the background contribution subtracted. The background contribution was determined by measuring length changes in a piece of copper of the same dimensions as the NCSO crystal [see Fig. S4(b)]. In zero magnetic field, the relative length change  $(\Delta L/L_0)_{c^*}$  (with  $L_0$  defined as the  $c^*$ -axis sample length at  $T = 300$  K and 0 T) decreases upon cooling, showing an S-shaped anomaly around the Néel temperature  $T_N \sim 7$  K, as seen in Fig. 1(d), indicating that NCSO contracts along the  $c^*$  axis upon entering the magnetically ordered state, in contrast to  $\alpha$ - $\text{RuCl}_3$  where cooling across  $T_N$  leads to the sample expansion [44].

The cooling and warming curves show a perfect reversibility around  $T_N$ , suggesting the second-order nature of this transition (see Fig. S9). The corresponding linear thermal expansion coefficient,  $\alpha = \frac{1}{L_0} \frac{d\Delta L}{dT}$ , shown on the right y axis of Fig. 1(d), reveals a pronounced positive peak that signals the onset of the AFM order. The substantial lattice changes  $[(\Delta L/L_0)_{c^*} \sim 6 \times 10^{-5}]$  observed around  $T_N$  suggest the presence of magnetoelastic coupling in NCSO. For comparison, we have also included the zero-field relative length changes of various honeycomb magnets as shown in Table S1 of the Supplemental Material Ref. [37].

Next, we study the effect of an applied magnetic field on the AFM order for both in-plane field directions (see Fig. 2).

At low fields, the length changes are similar to those in zero field, but as the magnetic field approaches  $B_{c2}$ , the responses in the two orientations diverge significantly. For  $B \parallel a$ , the S-shaped anomaly in  $(\Delta L/L_0)_{c^*}$  at  $T_N$  evolves into a broad maximum around the critical field  $B_{c2} \sim 1.8$  T, and at higher fields the relative length change increases upon cooling, indicating a lattice expansion. In contrast, for  $B \parallel b$ ,  $(\Delta L/L_0)_{c^*}$  continues to decrease smoothly toward low temperatures for fields above  $B_{c2} \sim 0.85$  T, indicating that the  $c^*$ -axis length keeps shrinking for this field configuration. To track these field-induced anomalies, we plot the corresponding values of  $\alpha_{c^*}$  in Figs. 2(b) and 2(d) for  $B \parallel a$  and  $B \parallel b$ , respectively. With increasing magnetic field, the sharp peak in  $\alpha_{c^*}$  shifts to lower temperatures, reflecting the suppression of AFM order, consistent with the trends observed in dc magnetic susceptibility and specific heat (Figs. S2 and S5) [37]. Near  $B_{c2}$ , the peak of  $\alpha_{c^*}$  broadens significantly, and a distinct behavior emerges beyond  $B_{c2}$ : while a broad maximum is seen for both field orientations, the absolute values of  $\alpha_{c^*}$  differ notably. For  $B \parallel b$ ,  $\alpha_{c^*}$  shows a broad maximum with the positive values, but for  $B \parallel a$ ,  $\alpha_{c^*}$  becomes negative, and eventually a minimum in  $\alpha_{c^*}$  is observed. Similar type of a sign change in  $\alpha_c$  has been also reported in  $\text{Na}_2\text{Co}_2\text{TeO}_6$  [14]. The broad maximum (minimum) in  $\alpha_{c^*}$  observed above  $B_{c2}$  marks a crossover from the field-induced saturated state into the paramagnetic state. The contrasting behaviors observed for the two in-plane directions highlight the strong in-plane anisotropy of the magnetoelastic coupling in NCSO. The temperature values of the sharp anomalies observed in the ordered state and the positions of the broad maxima and minima in the

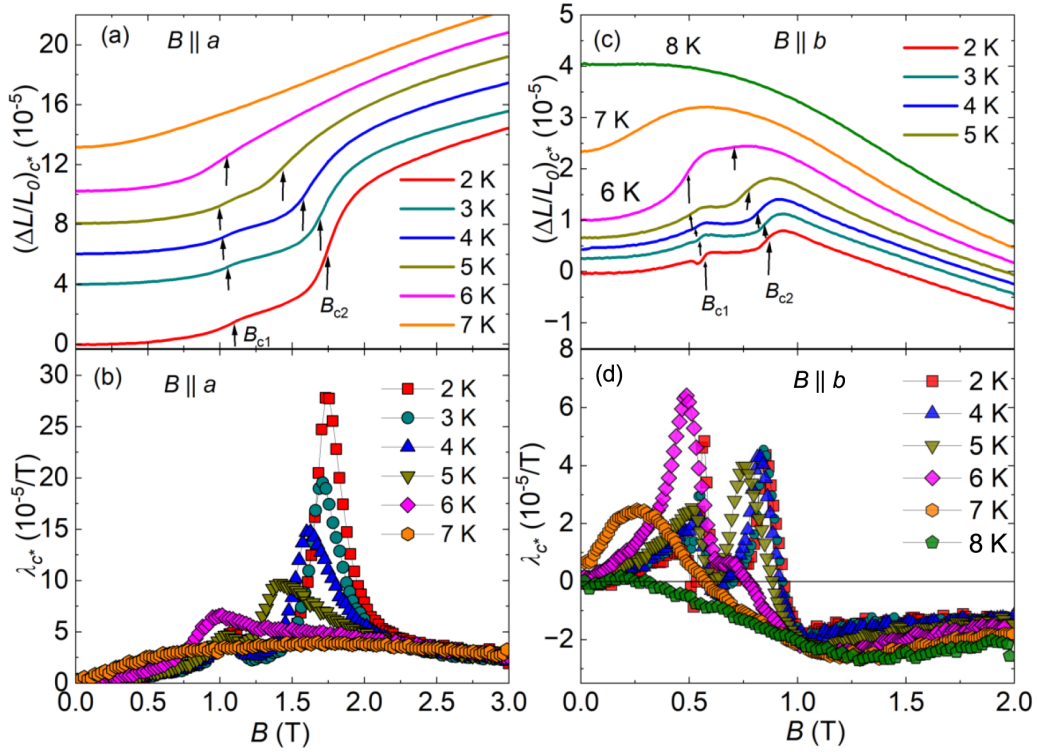


FIG. 3. (a), (c) Field dependence of relative length changes at constant temperature for  $B \parallel a$  and  $B \parallel b$ , respectively (the data are measured upon increasing the field). The curves are shifted vertically for clarity. The arrows indicate the positions of  $B_{c1}$  and  $B_{c2}$ . (b), (d) Their corresponding coefficients of magnetostriction vs field.

field-polarized state are compiled in the  $B$ - $T$  phase diagram shown in Fig. 7.

### B. Magnetostriction

To monitor the field-induced changes in NCSO, we measured magnetostriction (MS) at constant temperature over the temperature range  $2 \text{ K} \leq T \leq 8 \text{ K}$  (Fig. 3). For  $T \leq 5 \text{ K}$ , the  $c^*$ -axis magnetostriction shows a clear field-orientation dependence. For  $B \parallel a$ ,  $(\Delta L/L_0)_{c^*}$  increases monotonically with field, with distinct slope changes at  $B_{c1}$  and  $B_{c2}$ . In contrast, for  $B \parallel b$ ,  $(\Delta L/L_0)_{c^*}$  increases only up to  $B_{c1}$ , after which it becomes nonmonotonic, with clear anomalies at  $B_{c1}$  and  $B_{c2}$ . In the high-temperature regime above 6 K (7 K), the length change becomes featureless for  $B \parallel a$  ( $B \parallel b$ ). Similar to the length change measured at constant field, the field-dependent length changes also show contrasting behaviors around  $B_{c2}$  for the  $a$  and  $b$  directions. While  $(\Delta L/L_0)_{c^*}$  continues to increase beyond 1.8 T for  $B \parallel a$ , it shrinks for  $B \parallel b$  at all measured temperatures. Notably, the length change for  $B \parallel a$  is nearly ten times larger than that for  $B \parallel b$ , indicating a more significant magnetoelastic coupling when field is applied along  $a$ .

The corresponding coefficients of the linear magnetostriction ( $\lambda = \frac{1}{L_0} \frac{d\Delta L}{dB}$ ) are plotted in Figs. 3(b) and 3(d) for both in-plane directions. At temperatures below 5 K, two sharp anomalies at  $B_{c1}$  and  $B_{c2}$  are clearly evident as distinct peaks in the  $\lambda_{c^*}$  versus  $B$  curves, whereas no additional anomaly is observed below  $B_{c1}$ . Such a low-field anomaly has been seen in various field-dependent thermodynamic measurements on  $\alpha$ - $\text{RuCl}_3$  [36,44,45] where it is usually interpreted as the domain repopulation within the zigzag state. The absence of this

anomaly in NCSO corroborates the monodomain double- $q$  ground state of this material.

The field hysteresis at  $B_{c1}$  (see Figs. S6 and S7) suggests the first-order nature of this transition. The hysteresis persists up to 4 K (5 K) for the  $a$  ( $b$ ) direction, and disappears upon approaching  $T_N$ . Conversely, the up  $\leftrightarrow$  down data at  $B_{c2}$  are reversible at all measured temperatures. Above 5 K (6 K) with  $B \parallel a$  ( $B \parallel b$ ), the two transitions merge, whereas at even higher temperatures  $\lambda(B)$  becomes featureless. The corresponding critical fields for each temperature are consistent with the results obtained from the  $M(B)$  measurements, as shown in Fig. 7. We also find a close similarity between field dependencies of  $\lambda$  and  $dM/dB$ , as shown in Figs. S6 and S7.

### C. Effect of out-of-plane strain

We now explore the origin of anisotropic magnetostriction using DFT +  $U$  + SO calculations. To this end, we relaxed atomic positions and lattice parameters of NCSO for fixed values of the interlayer distance  $c^*$  assuming different spin configurations: the ferromagnetic state with the spins pointing along either  $a$  or  $b$ , as well as the zigzag and double- $q$  states as possible magnetic structures in zero field. Figures 4(a) and 4(b) display the parabolic dependence of total energy on  $c^*$ , as expected for the elastic strain applied to the material. The slope of these curves returns an effective elastic constant of 230 GPa, which is within the expected range for an ionic compound.

Remarkably, different spin configurations lead to different equilibrium values of  $c^*$ . Their hierarchy follows our

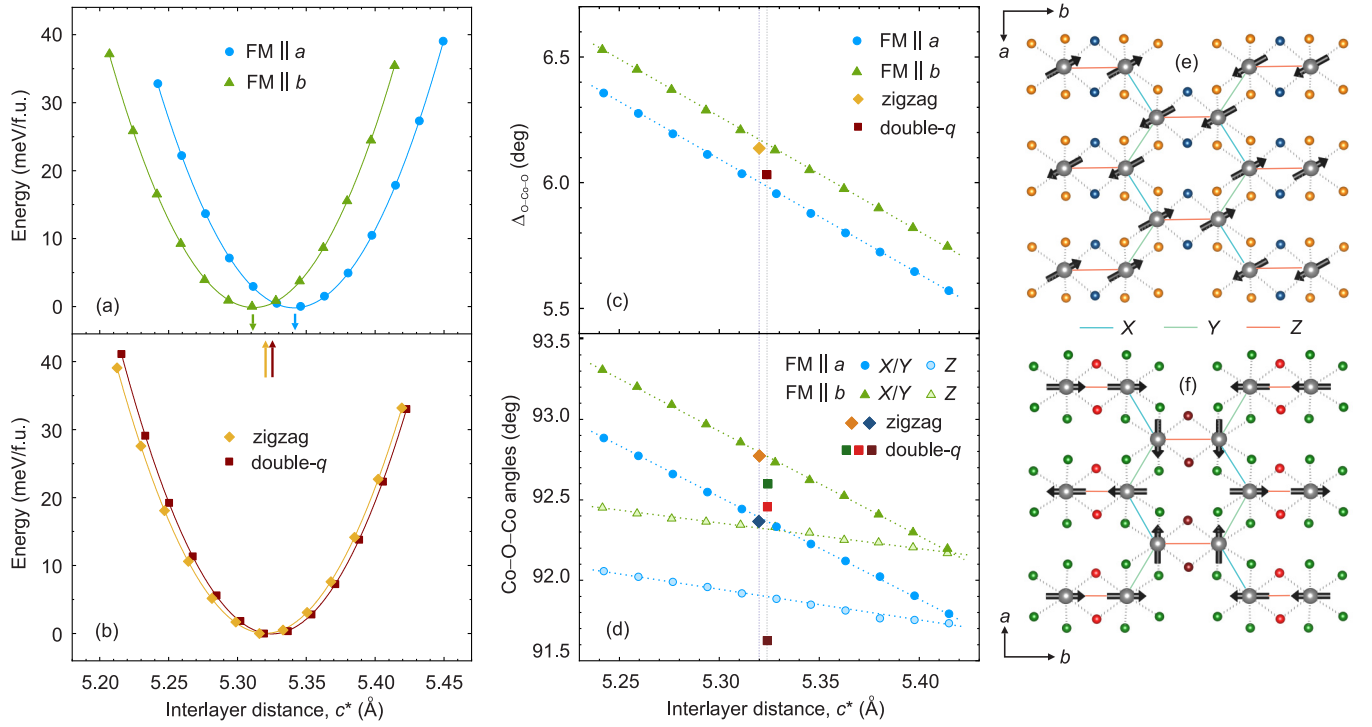


FIG. 4. (a), (b) Total energy depending on the interlayer distance  $c^*$  for different spin configurations of NCSO. The positions of the energy minima are indicated by arrows. (c) Deformation of the  $\text{CoO}_6$  octahedra gauged by  $\Delta_{\text{O-Co-O}}$ , the average deviation of O-Co-O angles from the median value of  $90.0^\circ$ . (d) Co-O-Co bond angles. The color codes for the zigzag and double- $q$  states correspond to the different oxygen atoms, as depicted in (e) and (f), respectively. Only the bond angles at the position of the energy minimum are shown in each case.

experimental results, namely, the interlayer distance in either the zigzag or double- $q$  state is shorter compared to the FM state for  $B \parallel a$  and longer than in the FM state for  $B \parallel b$ . The relative changes in  $c^*$  are somewhat larger than the values of  $(\Delta L/L_0)_{c^*} \simeq 1 \times 10^{-4}$  observed experimentally. This is probably an artifact of DFT +  $U$  + SO where  $\text{Co}^{2+}$  is treated as a spin- $\frac{3}{2}$  ion and not as the  $j_{\text{eff}} = \frac{1}{2}$  ion, leading to overestimated magnetoelastic energies, with somewhat exaggerated changes in  $c^*$  between the different spin configurations. Importantly, though, the trends in  $c^*$  are reproduced, and the effect of spin configurations on individual structural parameters can be explored in detail.

The difference in  $c^*$  between the FM states polarized along  $a$  and  $b$  could be related either to single-ion physics of  $\text{Co}^{2+}$ , or to anisotropic magnetic couplings. These effects are controlled by the local geometry of the  $\text{CoO}_6$  octahedra and the Co-O-Co bond angles, respectively. The monoclinic crystal structure of NCSO gives rise to a rather complex deformation of the  $\text{CoO}_6$  octahedra, but the median O-Co-O angle remains very close to  $90.0^\circ$  for all strains. Therefore, we quantify the octahedral deformation by the average absolute deviation of the O-Co-O angles from this median value. Figure 4(c) shows that the tensile strain along  $c^*$  renders the octahedra more symmetric, but for a given  $c^*$ , the difference in the deformation between the FM states polarized along  $a$  and  $b$  is about  $0.15^\circ$  only.

The Co-O-Co bond angles in Fig. 4(d) show a much larger difference of about  $0.4^\circ$  depending on the spin direction. This effect is a fingerprint of anisotropic exchange interactions because isotropic Heisenberg terms are insensitive to the di-

rection of spin. The presence of anisotropic exchange is in agreement with the earlier findings of either Kitaev [46,47] or off-diagonal anisotropy [35] terms being dominant in NCSO.

The monoclinic structure of NCSO features two distinct Co-O-Co bond angles that correspond to the X/Y and Z bonds in the Kitaev frame. The Co-O-Co angles for the Z bonds are systematically lower than those for the X/Y bonds. While both angles decrease with increasing  $c^*$ , they change at a different rate and approach each other at about 2% of the tensile strain. It is further instructive to compare these angles with the optimized values obtained for the zigzag and double- $q$  states [Fig. 4(d)]. The zigzag state involves FM spin-spin correlations on the Z bond and on one of the X, Y bonds. From Goodenough-Kanamori-Anderson rules, one expects that lower Co-O-Co angles should favor a FM coupling. Indeed, the respective angle is about  $92.4^\circ$  for the Z bond, but the second FM correlation has to set in on the Y bond with the larger angle. The double- $q$  state is accompanied by a much stronger structural deformation. The FM correlations are now confined to the Z bonds, and half of them, those with the spins along  $a$ , develop much smaller bond angles of  $91.6^\circ$ , which are comparable to the Z-bond angles in the FM state polarized along  $a$ . Altogether, one sees consistent changes in the Co-O-Co bond angles with respect to both spin direction and the type of magnetic order on each bond.

#### D. Magnetic Grüneisen parameter

We now return to the field-induced phase transitions in NCSO and study them in more detail using magnetocaloric

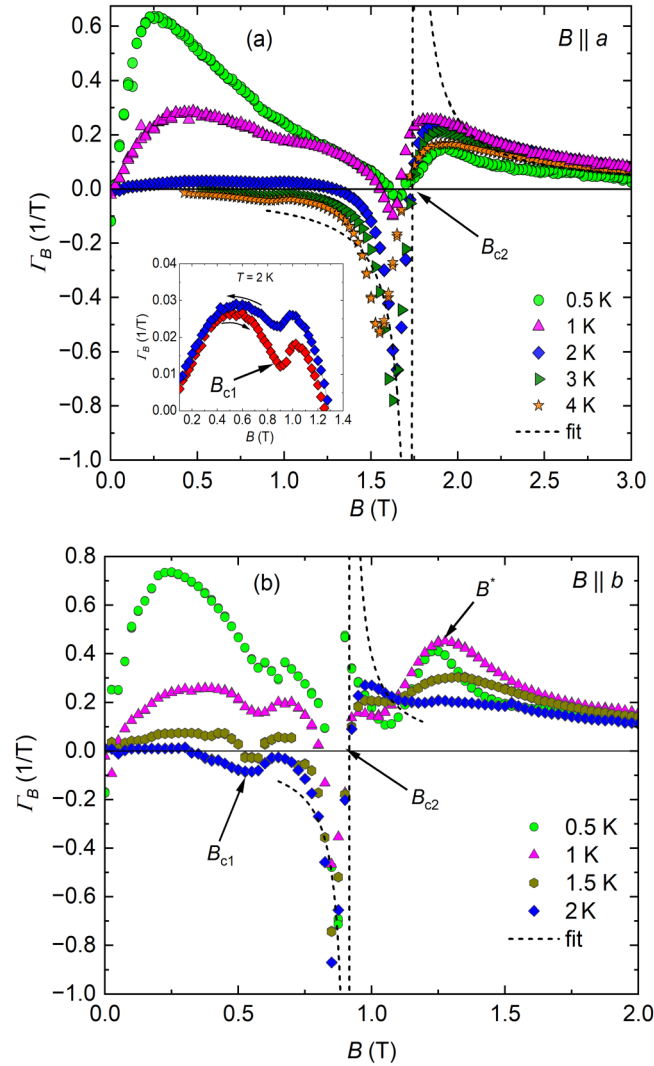


FIG. 5. Magnetic field dependence of  $\Gamma_B$  for (a)  $B \parallel a$  and (b)  $B \parallel b$ . The inset in (a) shows the hysteresis around  $B_{c1}$ . The dashed lines are the fit as described in the text.

effect (MCE) measured below the Néel temperature (Fig. 5). The magnetic Grüneisen parameter [ $\Gamma_B = (1/T)(\partial T/\partial B)_S$ ] which equals the relative adiabatic magnetocaloric effect displays anomalies at  $B_{c1}$  and  $B_{c2}$ . For both field directions, the anomaly at  $B_{c1}$  appears as a broad minimum, which is more pronounced for  $B \parallel b$ . Upon approaching  $B_{c2}$ , the shape of  $\Gamma_B$  changes abruptly and shows a divergent behavior with a sign change. The up-down sweep performed for  $B \parallel a$  reveals a pronounced field hysteresis around  $B_{c1}$ , but essentially no hysteresis near  $B_{c2}$ , in agreement with our magnetostriction and magnetization data.

For  $B \parallel a$ , several experimental results point to unusual behavior in the vicinity of  $B_{c2}$ . The NMR spin-lattice relaxation rate shows an enhancement of low-energy spin fluctuations together with a gapped regime around  $B_{c2}$  [31]. Thermal transport measurements likewise report a pronounced minimum in the thermal conductivity  $\kappa$  at  $B_{c2}$ , attributed to an enhanced phonon scattering by magnetic fluctuations [33]. In addition, ac-magnetostriction experiments identify a tricritical point close to  $B_{c2}$  [34]. While these observations raise the

possibility of the field-induced quantum criticality, they do not establish it conclusively. In our data, the field dependence of  $\Gamma_B(B)$  (Fig. 5) shows a divergence near  $B_{c2}$ , reminiscent of field-induced quantum critical points in itinerant magnets [48,49]. The sign change of  $\alpha_{c^*}$  at  $B_{c2}$  [Fig. 2(b)] is also consistent with such a scenario, although it is not a direct proof of quantum criticality.

To assess whether a quantum-critical description is applicable, we therefore fitted the field dependence of  $\Gamma_B$  near  $B_{c2}$  using a power law of the form  $\Gamma_B = G/(B - B_c)$ , where  $G$  is a constant [50]. In these fits,  $B_{c2}$  was fixed to the zero crossing values in  $\Gamma_B$ . The dashed lines in Fig. 5 show the corresponding fits, yielding the prefactors  $G$  of about 0.07 and 0.03 for  $B \parallel a$  and  $B \parallel b$ , respectively. These prefactors are much smaller compared to systems with established field-induced quantum criticality [48,49]. In addition, the apparent divergence of  $\Gamma_B$  becomes weaker on cooling, whereas for a genuine quantum-critical point it should be enhanced at low temperatures. Taken together, the small prefactors and the temperature evolution of the  $\Gamma_B$  divergence suggest the absence of quantum-critical behavior in NCSO at  $B_{c2}$ .

Extending thermodynamic measurements on NCSO below 2 K not only evaluates quantum criticality in NCSO, but also exposes some further unusual features of this material. Figure 5 shows that  $\Gamma_B$  is small below  $B_{c1}$  at 2 K, but at lower temperatures it increases significantly and develops a broad feature around 0.3 T. Since this feature is not observed at higher temperatures, its origin is probably unrelated to domain repopulation. The monodomain double- $q$  ground state of NCSO further renders such a scenario unlikely. In order to shed more light on the behavior of NCSO at sub-Kelvin temperatures, we turn to the magnetization measurements below 2 K.

### E. Magnetization below 2 K

Figure 6 shows the field-dependent magnetization  $M(B)$  of NCSO at  $T = 0.4$  K (the data for other temperatures are shown in Fig. S3). As seen in Fig. 6, the magnetization undergoes abrupt changes with the field. It exhibits several clear steps that are especially pronounced for  $B \parallel b$ . Although the number of steps is relatively small, each step is well defined (sharp peaks in  $dM/dB$ ), thus pointing to abrupt changes of the spin configuration rather than a gradual canting of the magnetic moments.

Similar steplike behavior has been reported in other Co-based magnets. In  $\text{BaCo}_2(\text{AsO}_4)_2$ , field-dependent magnetization measurements at  $T = 0.14$  K reveal a series of small steps [51]. These have been interpreted in terms of slow domain and defect dynamics and avalanchelike jumps between metastable states of a frustrated magnet. A very similar behavior is also found in  $\text{BaCo}_2(\text{PO}_4)_2$  [52], where magnetostriction below 1 K shows jumps and tiny plateaus. Our data for NCSO, although showing fewer and more widely spaced steps, point in the same general direction.

Two further features are worth mentioning. First, the  $B_{c1}$  transition at 0.4 K is accompanied by a large and abrupt change in the magnetization for  $B \parallel b$  but not for  $B \parallel a$ . This difference reinforces the idea of Ref. [35] on the different nature of the field-induced state between  $B_{c1}$  and  $B_{c2}$  for

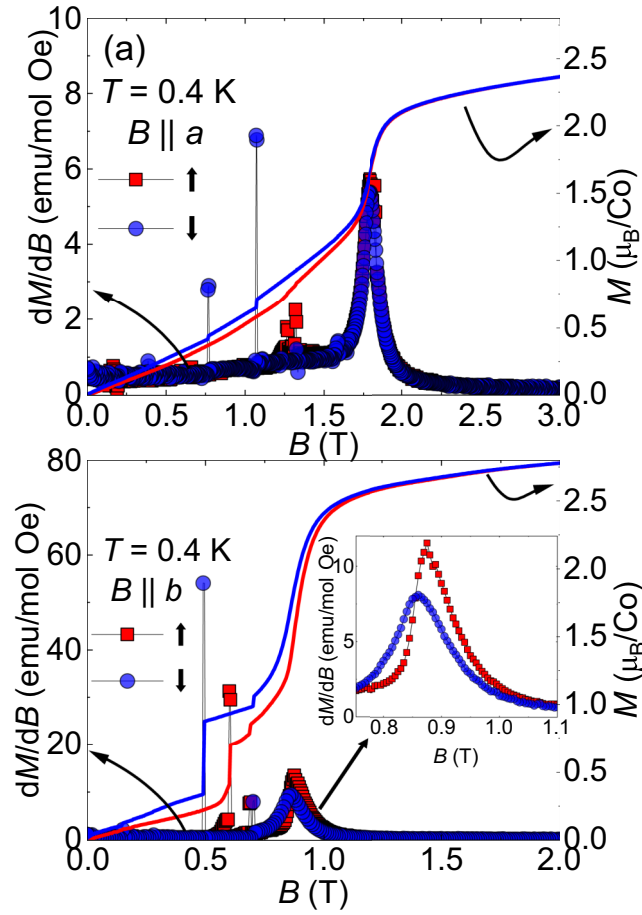


FIG. 6. Magnetization isotherms measured at  $T = 0.4$  K for (a)  $B \parallel a$  and (b)  $B \parallel b$  directions. The inset in (b) shows the tiny hysteresis observed around  $B_{c2}$ .

the two field directions,  $\frac{1}{3}$ -AFM for  $B \parallel a$  vs canted zigzag for  $B \parallel b$ , although the relation of these states to the size of the magnetization steps remains to be established. Second, all measurements above 2 K consistently show second-order nature of the transition at  $B_{c2}$ , but a small hysteresis appears in the  $M(B)$  data for  $B \parallel b$  [see inset of Fig. 6(b)]. This feature indicates that the second-order transition at  $B_{c2}$  becomes first order at sub-Kelvin temperatures and further excludes the presence of a quantum-critical point at  $B_{c2}$ .

#### IV. PHASE DIAGRAM AND DISCUSSION

Figure 7 shows the field-temperature phase diagram of NCSO, constructed from all our measurements discussed in the previous section. The phase boundaries are determined from the anomalies in the magnetization, specific heat, and dilatometry, namely, from the peaks in  $d(\chi T)/dT$ ,  $dM(B)/dB$ ,  $\alpha_{c^*}(T)$ ,  $\lambda_{c^*}(T)$ , and  $C_p(T)$ . For the MCE data, we take  $B_{c1}$  as the position of the broad minimum in  $\Gamma_B$ , and identify  $B_{c2}$  by the zero crossings of  $\Gamma_B$ . Above  $B_{c2}$ ,  $\alpha_{c^*}(T)$  exhibits a broad minimum (maximum) for  $B \parallel a$  ( $B \parallel b$ ); the position of this extremum at each field is used to mark the crossover between the high-temperature paramagnetic regime and the field-induced saturated state. Two magnetically or-

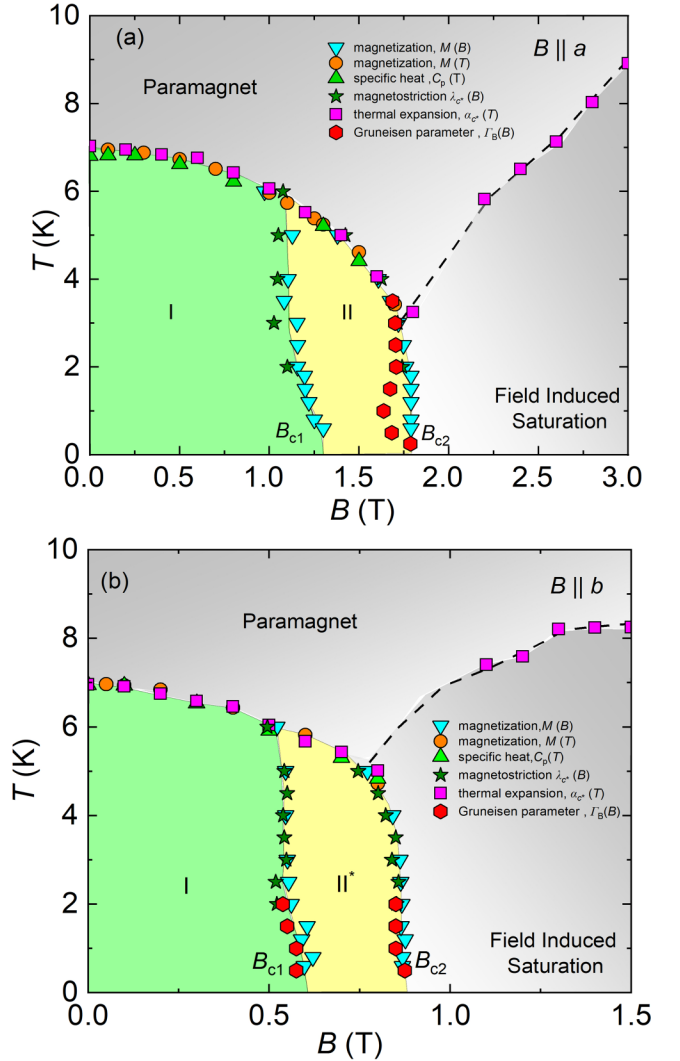


FIG. 7. Field-temperature phase diagram of NCSO constructed from various thermodynamic measurements for (a)  $B \parallel a$ , (b)  $B \parallel b$  directions. The anomalies in the  $\Gamma_B$  points in (a) may be due to the first-order nature of the transition at  $B_{c2}$  below 2 K.

dered states are identified for each field direction. Phase I entails the double- $q$  order according to neutron diffraction experiments [29], whereas phase II is described as the AFM- $\frac{1}{3}$  state for  $B \parallel a$  [25] and canted zigzag state for  $B \parallel b$  [35]. Our phase boundaries agree well with those reported previously above 2 K. They extend toward sub-Kelvin temperatures almost vertically, the main difference being the change in the nature of the transition at  $B_{c2}$  from second order toward first order.

The initial pressure dependence of the magnetic transitions can be inferred from the thermal expansion and magnetostriction data. The zero-field transition at  $T_N \simeq 7$  K is second order (see Fig. S9). For such a transition, the uniaxial pressure dependence of  $T_N$  along the  $c^*$  axis can be estimated using the Ehrenfest relation  $dT_N/dp_{c^*} = V_m T_N \Delta\alpha_{c^*} / \Delta C_p$ , where  $V_m = 8.04 \times 10^{-5} \text{ m}^3/\text{mol}$  is the molar volume,  $\Delta\alpha_{c^*}$  is the jump in the linear TE coefficient at  $T_N$ , and  $\Delta C_p$  is the specific-heat change at the transition. Using  $T_N \simeq 7$  K,  $\Delta\alpha_{c^*} = 2.5 \times 10^{-5} \text{ K}^{-1}$ , and  $\Delta C_p = 18 \text{ J/mol K}$ , we obtain

$dT_N/dp_{c^*} \simeq 0.77$  K/GPa, indicating that  $T_N$  should increase if pressure is applied along the  $c^*$  axis. Further insight into the pressure response follows from the Maxwell relation  $\lambda V = -(dM/dp)_{T,B}$ , where  $\lambda$  is the magnetostriction coefficient,  $V$  is the sample volume, and  $M$  is the magnetization. Although our measurements are limited to linear magnetostriction along  $c^*$ , rather than the full volume magnetostriction, the positive  $\lambda_{c^*}$  implies  $(dM/dp)_{T,B} < 0$ . Hence, the magnetization is expected to decrease upon compression along the  $c^*$  axis of NCsO.

Now, we discuss the region close to  $B_{c2}$ , which is of particular interest in the context of possible field-induced QSL. The behavior of  $\alpha$ -RuCl<sub>3</sub> in this region is still vividly debated [10]. In the case of BaCo<sub>2</sub>(AsO<sub>4</sub>)<sub>2</sub>, the formation of a gapless QSL has been proposed on the basis of thermal conductivity measurements [16], but the observation of persistent magnetic Bragg peaks in this region [15] argues against this scenario. A similar discussion exists for Na<sub>2</sub>Co<sub>2</sub>TeO<sub>6</sub>, where it is also debated whether a QSL phase appears above  $B_{c2}$  [13]. In NCsO, the in-plane anisotropy renders the behavior above  $B_{c2}$  strongly dependent on the field direction.

For  $B \parallel a$ , NMR measurements indicated a possible additional phase extending from  $B_{c2}$  up to 3.0 T according to the nonmonotonic evolution of the spin gap [31], but our data clearly exclude any phase transitions beyond  $B_{c2}$ . A more plausible scenario is the crossover from second-order toward first-order transition upon cooling and phase coexistence around  $B_{c2}$ , which would also be consistent with the observation of magnetic Bragg peaks even above  $B_{c2}$  [25].

The  $B \parallel b$  case is more intriguing, because  $\Gamma_B$  shows a peak at 1.2–1.3 T, well above  $B_{c2}$  (Fig. 5). This feature is reminiscent of the shoulder in  $\Gamma_B$  reported in  $\alpha$ -RuCl<sub>3</sub> in the field range of the putative QSL [53], but that shoulder can be understood in terms of a level crossing of two low-lying excited states and does not unambiguously signal a QSL [45]. A similar explanation may apply to NCsO too. Reference [35] further reports a shoulderlike anomaly in the specific heat for  $B \parallel b$ , which was discussed in terms of short-range magnetic order. Another

important observation is that the shoulder in  $\Gamma_B$  appears below 2 K only, concurrent with the peak in  $\Gamma_B$  around 0.3 T and the steps in the magnetization. All these features may be potentially related to slow dynamics, possibly of domain boundaries, and clearly deserve further investigation.

## V. SUMMARY

In summary, we have investigated the thermodynamic properties of the honeycomb magnet Na<sub>3</sub>Co<sub>2</sub>SbO<sub>6</sub> using high-resolution dilatometry, magnetocaloric effect, magnetization, and specific-heat measurements and extended the phase diagram of this material into the sub-Kelvin temperature range. In the ordered state, two field-induced transitions at  $B_{c1}$  and  $B_{c2}$  are observed in all probes. Whereas the former transition is first order, the latter transition is second order above 2 K and gradually becomes first order at lower temperatures. Anisotropic magnetoelastic coupling is reproduced using *ab initio* calculations and interpreted as an effect of changes in the Co–O–Co bond angles caused by the out-of-plane strain. Our data rule out quantum critical behavior near  $B_{c2}$  and further show intriguing steplike features in the magnetization, which are characteristic of metastable states. There is no thermodynamic evidence for a field-induced QSL state in Na<sub>3</sub>Co<sub>2</sub>SbO<sub>6</sub>.

## ACKNOWLEDGMENTS

This work was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation), TRR 360, Grant No. 492547816 (subprojects B1 and B2). The authors gratefully acknowledge the use of computing resources of the ALCC HPC cluster (Institute of Physics, University of Augsburg).

## DATA AVAILABILITY

The data that support the findings of this article are openly available [54], embargo periods may apply.

- 
- [1] A. Kitaev, Anyons in an exactly solved model and beyond, *Ann. Phys.* **321**, 2 (2006).
  - [2] S. M. Winter, A. A. Tsirlin, M. Daghofer, J. van den Brink, Y. Singh, P. Gegenwart, and R. Valentí, Models and materials for generalized Kitaev magnetism, *J. Phys.: Condens. Matter* **29**, 493002 (2017).
  - [3] B. Shen, A. Jesche, M. L. Seidler, F. Freund, P. Gegenwart, and A. A. Tsirlin, Interplay of magnetism and dimerization in the pressurized Kitaev material  $\beta$ -Li<sub>2</sub>IrO<sub>3</sub>, *Phys. Rev. B* **104**, 134426 (2021).
  - [4] P. Lampen-Kelley, A. Banerjee, A. A. Aczel, H. B. Cao, M. B. Stone, C. A. Bridges, J.-Q. Yan, S. E. Nagler, and D. Mandrus, Destabilization of magnetic order in a dilute Kitaev spin liquid candidate, *Phys. Rev. Lett.* **119**, 237203 (2017).
  - [5] A. Banerjee, P. Lampen-Kelley, J. Knolle, C. Balz, A. A. Aczel, B. Winn, Y. Liu, D. Pajerowski, J. Yan, C. A. Bridges, A. T. Savici, B. C. Chakoumakos, M. D. Lumsden, D. A. Tennant, R. Moessner, D. G. Mandrus, and S. E. Nagler, Excitations in the field-induced quantum spin liquid state of  $\alpha$ -RuCl<sub>3</sub>, *npj Quantum Mater.* **3**, 8 (2018).
  - [6] H. Liu and G. Khaliullin, Pseudospin exchange interactions in  $d^7$  cobalt compounds: Possible realization of the Kitaev model, *Phys. Rev. B* **97**, 014407 (2018).
  - [7] R. Sano, Y. Kato, and Y. Motome, Kitaev-Heisenberg Hamiltonian for high-spin  $d^7$  Mott insulators, *Phys. Rev. B* **97**, 014408 (2018).
  - [8] S. M. Winter, Magnetic couplings in edge-sharing high-spin  $d^7$  compounds, *J. Phys. Mater.* **5**, 045003 (2022).
  - [9] X. Liu and H.-Y. Kee, Non-Kitaev versus Kitaev honeycomb cobaltates, *Phys. Rev. B* **107**, 054420 (2023).
  - [10] S. Kim, B. Yuan, and Y.-J. Kim,  $\alpha$ -RuCl<sub>3</sub> and other Kitaev materials, *APL Mater.* **10**, 080903 (2022).

- [11] R. Zhong, T. Gao, N. P. Ong, and R. J. Cava, Weak-field induced nonmagnetic state in a Co-based honeycomb, *Sci. Adv.* **6**, eaay6953 (2020).
- [12] S. Lee, S. Zhang, S. M. Thomas, L. Pressley, C. A. Bridges, E. S. Choi, V. S. Zapf, S. M. Winter, and M. Lee, Quantum order by disorder is a key to understanding the magnetic phases of  $\text{BaCo}_2(\text{AsO}_4)_2$ , *npj Quantum Mater.* **10**, 11 (2025).
- [13] S. Fang, K. Imamura, Y. Mizukami, R. Namba, K. Ishihara, K. Hashimoto, and T. Shibauchi, Field-angle-resolved specific heat in  $\text{Na}_2\text{Co}_2\text{TeO}_6$ : Evidence against Kitaev quantum spin liquid, *Phys. Rev. Lett.* **134**, 106701 (2025).
- [14] J. Arneth, K.-Y. Choi, R. Kalaivanan, R. Sankar, and R. Klingeler, Signatures of a quantum critical endpoint in the Kitaev candidate  $\text{Na}_2\text{Co}_2\text{TeO}_6$ , *Phys. Rev. B* **110**, L140402 (2024).
- [15] P. K. Mukharjee, B. Shen, S. Erdmann, A. Jesche, J. Kaiser, P. R. Baral, O. Zaharko, P. Gegenwart, and A. A. Tsirlin, Intermediate field-induced phase of the honeycomb magnet  $\text{BaCo}_2(\text{AsO}_4)_2$ , *Phys. Rev. B* **110**, L140407 (2024).
- [16] C. Tu, D. Dai, X. Zhang, C. Zhao, X. Jin, B. Gao, T. Chen, P. Dai, and S. Li, Evidence for mobile gapless spinons in a honeycomb lattice, *Chin. Phys. Lett.* **42**, 067304 (2025).
- [17] X. Hong, M. Gillig, R. Hentrich, W. Yao, V. Kocsis, A. R. Witte, T. Schreiner, D. Baumann, N. Pérez, A. U. B. Wolter, Y. Li, B. Büchner, and C. Hess, Strongly scattered phonon heat transport of the candidate Kitaev material  $\text{Na}_2\text{Co}_2\text{TeO}_6$ , *Phys. Rev. B* **104**, 144426 (2021).
- [18] S. Guang, N. Li, R. L. Luo, Q. Huang, Y. Wang, X. Yue, K. Xia, Q. Li, X. Zhao, G. Chen, H. Zhou, and X. Sun, Thermal transport of fractionalized antiferromagnetic and field-induced states in the Kitaev material  $\text{Na}_2\text{Co}_2\text{TeO}_6$ , *Phys. Rev. B* **107**, 184423 (2023).
- [19] S. Zhang, S. Lee, A. J. Woods, W. K. Peria, S. M. Thomas, R. Movshovich, E. Brosha, Q. Huang, H. Zhou, V. S. Zapf, and M. Lee, Electronic and magnetic phase diagrams of the Kitaev quantum spin liquid candidate  $\text{Na}_2\text{Co}_2\text{TeO}_6$ , *Phys. Rev. B* **108**, 064421 (2023).
- [20] P. Pilch, L. Peedu, A. K. Bera, S. M. Yusuf, U. Nagel, T. Röm, and Z. Wang, Field- and polarization-dependent quantum spin dynamics in the honeycomb magnet  $\text{Na}_2\text{Co}_2\text{TeO}_6$ : Magnetic excitations and continuum, *Phys. Rev. B* **108**, L140406 (2023).
- [21] W. Yao, Y. Zhao, Y. Qiu, C. Balz, J. R. Stewart, J. W. Lynn, and Y. Li, Magnetic ground state of the Kitaev  $\text{Na}_2\text{Co}_2\text{TeO}_6$  spin liquid candidate, *Phys. Rev. Res.* **5**, L022045 (2023).
- [22] A. K. Bera, S. M. Yusuf, F. Orlandi, P. Manuel, L. Bhaskaran, and S. A. Zvyagin, Field-induced phase transitions and anisotropic magnetic properties of the Kitaev-Heisenberg compound  $\text{Na}_2\text{Co}_2\text{TeO}_6$ , *Phys. Rev. B* **108**, 214419 (2023).
- [23] L. Shi, X. Li, R. Li, Y. Li, T. Dong, J. Luo, X. Wang, and N. Wang, Field and temperature evolution of the magnetic excitations in the field-induced state of  $\text{Na}_2\text{Co}_2\text{TeO}_6$ , *Phys. Rev. B* **112**, 184406 (2025).
- [24] Y. Gu, X. Jin, and Y. Li, On the multi- $q$  characteristics of magnetic ground states of honeycomb cobalt oxides, *Chin. Phys. Lett.* **42**, 027303 (2025).
- [25] X. Li, Y. Gu, Y. Chen, V. O. Garlea, K. Iida, K. Kamazawa, Y. Li, G. Deng, Q. Xiao, X. Zheng, Z. Ye, Y. Peng, I. A. Zaliznyak, J. M. Tranquada, and Y. Li, Giant magnetic in-plane anisotropy and competing instabilities in  $\text{Na}_3\text{Co}_2\text{SbO}_6$ , *Phys. Rev. X* **12**, 041024 (2022).
- [26] L. Viciu, Q. Huang, E. Morosan, H. Zandbergen, N. Greenbaum, T. McQueen, and R. Cava, Structure and basic magnetic properties of the honeycomb lattice compounds  $\text{Na}_2\text{Co}_2\text{TeO}_6$  and  $\text{Na}_3\text{Co}_2\text{SbO}_6$ , *J. Solid State Chem.* **180**, 1060 (2007).
- [27] C. Wong, M. Avdeev, and C. D. Ling, Zig-zag magnetic ordering in honeycomb-layered  $\text{Na}_3\text{Co}_2\text{SbO}_6$ , *J. Solid State Chem.* **243**, 18 (2016).
- [28] J.-Q. Yan, S. Okamoto, Y. Wu, Q. Zheng, H. D. Zhou, H. B. Cao, and M. A. McGuire, Magnetic order in single crystals of  $\text{Na}_3\text{Co}_2\text{SbO}_6$  with a honeycomb arrangement of  $3d^7$   $\text{Co}^{2+}$  ions, *Phys. Rev. Mater.* **3**, 074405 (2019).
- [29] Y. Gu, X. Li, Y. Chen, K. Iida, A. Nakao, K. Munakata, V. O. Garlea, Y. Li, G. Deng, I. A. Zaliznyak, J. M. Tranquada, and Y. Li, In-plane multi- $q$  magnetic ground state of  $\text{Na}_3\text{Co}_2\text{SbO}_6$ , *Phys. Rev. B* **109**, L060410 (2024).
- [30] P. Miao, X. Jin, W. Yao, Y. Chen, A. Koda, Z. Tan, W. Xie, W. Ji, T. Kamiyama, and Y. Li, Persistent spin dynamics in magnetically ordered honeycomb-lattice cobalt oxides, *Phys. Rev. B* **109**, 134431 (2024).
- [31] Z. Hu, Y. Chen, Y. Cui, S. Li, C. Li, X. Xu, Y. Chen, X. Li, Y. Gu, R. Yu, R. Zhou, Y. Li, and W. Yu, Field-induced phase transitions and quantum criticality in the honeycomb antiferromagnet  $\text{Na}_3\text{Co}_2\text{SbO}_6$ , *Phys. Rev. B* **109**, 054411 (2024).
- [32] R. Li, L. Shi, Y. Gu, Y. Chen, X. Li, Q. Wu, S. Xu, D. Wu, Y. Li, F. Wang, T. Dong, and N. Wang, Field-induced spin continuum in twin-free  $\text{Na}_3\text{Co}_2\text{SbO}_6$  revealed by magneto-THz spectroscopy, *Chin. Phys. Lett.* **42**, 120711 (2025).
- [33] H. Fan, Y. Chen, Y. Gu, X. Li, Y. Li, and X. Lin, Phonon heat transport and anisotropic tuning of quantum fluctuations in a frustrated honeycomb magnet, *Phys. Rev. B* **111**, 085119 (2025).
- [34] X. Mi, X. Li, L. Zhang, Y. Gu, A. Wang, Y. Li, Y. Chai, and M. He, Precisely tracking critical spin fluctuations using the AC magnetostriction coefficient: A case study on the Kitaev spin liquid candidate  $\text{Na}_3\text{Co}_2\text{SbO}_6$ , *Phys. Rev. B* **111**, 014417 (2025).
- [35] K. K. Bestha, M. Sahoo, N. Francini, R. Kluge, R. Morrow, A. Maljuk, S. Wurmehl, S. Luther, Y. Skourski, H. Kühne, S. Mishra, J. Geck, M. Brando, B. Büchner, L. T. Corredor, L. Janssen, and A. U. B. Wolter, Field-induced magnetic phases in the Kitaev candidate  $\text{Na}_3\text{Co}_2\text{SbO}_6$ , *Phys. Rev. B* **113**, L100412 (2026).
- [36] R. Schönmann, S. Imajo, F. Weickert, J. Yan, D. G. Mandrus, Y. Takano, E. L. Brosha, P. F. S. Rosa, S. E. Nagler, K. Kindo, and M. Jaime, Thermal and magnetoelastic properties of  $\alpha$ - $\text{RuCl}_3$  in the field-induced low-temperature states, *Phys. Rev. B* **102**, 214432 (2020).
- [37] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/tn1m-sjdh> for details of crystal growth, specific heat measurement, additional magnetization, thermal expansion, and magnetostriction measurements, including Refs. [14,15,25,28,44,52,55].
- [38] Y. Tokiwa and P. Gegenwart, High-resolution alternating-field technique to determine the magnetocaloric effect of metals

- down to very low temperatures, *Rev. Sci. Instrum.* **82**, 013905 (2011).
- [39] R. Küchler, A. Wörl, P. Gegenwart, M. Berben, B. Bryant, and S. Wiedmann, The world's smallest capacitive dilatometer, for high-resolution thermal expansion and magnetostriction in high magnetic fields, *Rev. Sci. Instrum.* **88**, 083903 (2017).
- [40] G. Kresse and J. Furthmüller, Efficiency of *ab-initio* total energy calculations for metals and semiconductors using a plane-wave basis set, *Comput. Mater. Sci.* **6**, 15 (1996).
- [41] G. Kresse and J. Furthmüller, Efficient iterative schemes for *ab initio* total-energy calculations using a plane-wave basis set, *Phys. Rev. B* **54**, 11169 (1996).
- [42] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized gradient approximation made simple, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [43] D. Szaller, L. Prodan, K. Geirhos, V. Felea, Y. Skourski, D. Gorbunov, T. Förster, T. Helm, T. Nomura, A. Miyata, S. Zherlitsyn, J. Wosnitzer, A. A. Tsirlin, V. Tsurkan, and I. Kézsmárki, Coexistence of antiferromagnetism and ferrimagnetism in adjacent honeycomb layers, *Phys. Rev. B* **111**, 184404 (2025).
- [44] S. Gass, P. M. Cönsoli, V. Kocsis, L. T. Corredor, P. Lampen-Kelley, D. G. Mandrus, S. E. Nagler, L. Janssen, M. Vojta, B. Büchner, and A. U. B. Wolter, Field-induced transitions in the Kitaev material  $\alpha$ -RuCl<sub>3</sub> probed by thermal expansion and magnetostriction, *Phys. Rev. B* **101**, 245158 (2020).
- [45] S. Bachus, D. A. S. Kaib, Y. Tokiwa, A. Jesche, V. Tsurkan, A. Loidl, S. M. Winter, A. A. Tsirlin, R. Valentí, and P. Gegenwart, Thermodynamic perspective on field-induced behavior of  $\alpha$ -RuCl<sub>3</sub>, *Phys. Rev. Lett.* **125**, 097203 (2020).
- [46] M. Songvilay, J. Robert, S. Petit, J. A. Rodríguez-Rivera, W. D. Ratcliff, F. Damay, V. Balédent, M. Jiménez-Ruiz, P. Lejay, E. Pachoud, A. Hadj-Azzem, V. Simonet, and C. Stock, Kitaev interactions in the Co honeycomb antiferromagnets Na<sub>3</sub>Co<sub>2</sub>SbO<sub>6</sub> and Na<sub>2</sub>Co<sub>2</sub>TeO<sub>6</sub>, *Phys. Rev. B* **102**, 224429 (2020).
- [47] A. L. Sanders, R. A. Mole, J. Liu, A. J. Brown, D. Yu, C. D. Ling, and S. Rachel, Dominant Kitaev interactions in the honeycomb materials Na<sub>3</sub>Co<sub>2</sub>SbO<sub>6</sub> and Na<sub>2</sub>Co<sub>2</sub>TeO<sub>6</sub>, *Phys. Rev. B* **106**, 014413 (2022).
- [48] Y. Tokiwa, M. Mchawat, R. S. Perry, and P. Gegenwart, Multiple metamagnetic quantum criticality in Sr<sub>3</sub>Ru<sub>2</sub>O<sub>7</sub>, *Phys. Rev. Lett.* **116**, 226402 (2016).
- [49] Y. Tokiwa, M. Garst, P. Gegenwart, S. L. Bud'ko, and P. C. Canfield, Quantum bicriticality in the heavy-fermion metamagnet YbAgGe, *Phys. Rev. Lett.* **111**, 116401 (2013).
- [50] M. Garst and A. Rosch, Sign change of the Grüneisen parameter and magnetocaloric effect near quantum critical points, *Phys. Rev. B* **72**, 205129 (2005).
- [51] A. Devillez, J. Robert, E. Lhotel, R. Ballou, C. Cavenel, F. Denis Romero, Q. Faure, H. Jacobsen, J. Lass, D. G. Mazzone, U. B. Hansen, M. Enderle, S. Raymond, S. De Brion, V. Simonet, and M. Songvilay, Bond-dependent interactions and ill-ordered state in the honeycomb cobaltate BaCo<sub>2</sub>(AsO<sub>4</sub>)<sub>2</sub>, *Phys. Rev. Res.* **7**, L042040 (2025).
- [52] X. Wang, R. Sharma, P. Becker, L. Bohatý, and T. Lorenz, Single-crystal study of the honeycomb XXZ magnet BaCo<sub>2</sub>(PO<sub>4</sub>)<sub>2</sub> in magnetic fields, *Phys. Rev. Mater.* **7**, 024402 (2023).
- [53] S. Bachus, D. A. S. Kaib, A. Jesche, V. Tsurkan, A. Loidl, S. M. Winter, A. A. Tsirlin, R. Valentí, and P. Gegenwart, Angle-dependent thermodynamics of  $\alpha$ -RuCl<sub>3</sub>, *Phys. Rev. B* **103**, 054440 (2021).
- [54] P. K. Mukharjee, S. Erdmann, L. Wang, J. Kaiser, A. Jesche, P. Puphal, M. Isobe, M. Hepting, B. Keimer, P. Gegenwart, and A. A. Tsirlin, Anisotropic magnetoelastic coupling in the honeycomb magnet Na<sub>3</sub>Co<sub>2</sub>SbO<sub>6</sub> [Data set], Zenodo (2026), <https://doi.org/10.5281/zenodo.18795733>.
- [55] V. K. Anand and D. C. Johnston, Antiferromagnetism in EuCu<sub>2</sub>As<sub>2</sub> and EuCu<sub>1.82</sub>As<sub>2</sub> single crystals, *Phys. Rev. B* **91**, 184403 (2015).