

Field-induced magnetic phases in the Kitaev candidate $\text{Na}_3\text{Co}_2\text{SbO}_6$

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We report a rich anisotropic magnetic phase diagram of $\text{Na}_3\text{Co}_2\text{SbO}_6$, a previously proposed cobaltate Kitaev candidate, based on field- and temperature-dependent magnetization, specific heat, and magnetocaloric effect studies. At low temperatures, our experiments uncover a low-lying $j_{\text{eff}} = \frac{1}{2}$ state with an antiferromagnetic (AFM) ground state and pronounced in-plane versus out-of-plane anisotropy. The experimentally identified magnetic phases are theoretically characterized through classical Monte Carlo simulations within an extended Kitaev-Heisenberg model with additional ring exchange interactions. The resulting phase diagram reveals a variety of exotic field-induced magnetic phases, including double- $\mathbf{q}$, $\frac{1}{3}$ -AFM, zigzag, and vortex phases.

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Introduction. In frustrated magnets, the quest for the elusive quantum spin liquid [1] has driven the discovery of diverse magnetic phases. The interplay of multiple exchange interactions can give rise to unconventional collinear and noncollinear magnetic orders with topological spin structures, which hold potential for device applications [2]. In particular, materials hosting bond-dependent Kitaev exchange, in conjunction with other exchange interactions, can stabilize unique single- $\mathbf{q}$ and multi- $\mathbf{q}$ ground states, potentially leading to multiferroicity and the emergence of topological spin structures [3–7]. Here, honeycomb cobalt oxides containing high-spin Co^{2+} ions have attracted significant interest due to their ability to support strong Kitaev interactions alongside other symmetry-allowed exchange interactions [8,9].

Cobalt compounds with d^7 electronic configuration exhibit unique characteristics, including an exchange process driven by e_g electrons that cancels non-Kitaev interactions from

t_{2g} - t_{2g} channels, thereby enhancing the role of the Kitaev coupling in the case of neighboring octahedra sharing a common edge. Notably, the t_{2g} - e_g channel gives rise to a sizable Kitaev exchange, making cobaltates promising candidates to realize the various different states predicted in extended Kitaev-Heisenberg models [10–13]. The application of a magnetic field leads to additional exotic magnetic phases depending on the orientation and strength of the field [3,14,15]. Among the honeycomb cobaltates, $\text{Na}_3\text{Co}_2\text{SbO}_6$ is expected to exhibit a weaker trigonal field, allowing for bond-dependent anisotropic interactions alongside comparable non-Kitaev interactions. These conditions make it a promising candidate for realizing the extended Kitaev-Heisenberg model [16,17].

$\text{Na}_3\text{Co}_2\text{SbO}_6$ has recently garnered significant interest [18–29], particularly due to the predictions that tuning its trigonal crystal field via strain or pressure could bring it closer to the Kitaev spin liquid phase [10]. However, its antiferromagnetic (AFM) transition temperature is greatly influenced by the Na stoichiometry ($T_N \sim 4.3$ – 8.3 K) and, in the literature, the claimed ground state varies between zigzag and double $\mathbf{q}$ [27,30]. Notably, experimental strategies for distinguishing between single- and multi- $\mathbf{q}$ orders, along with their implications for microscopic model construction, are currently a topic of intense debate. This discussion extends not only to cobaltates [7,15,22,31–34] but also to other centrosymmetric compounds hosting three-dimensional topological spin structures [35,36]. Furthermore, in $\text{Na}_3\text{Co}_2\text{SbO}_6$, the exact nature of the field-induced phases remains under intense scrutiny, with magnetic phase diagrams yet to be fully understood, particularly for the out-of-plane field direction. Thermodynamic studies on polycrystalline samples suggest a field-induced spin-liquid-like phase [25], while AC composite

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magnetoelectric measurements, highly sensitive to spin fluctuations, reveal two field-induced tricritical points for in-plane magnetic fields, one of which appears to lie near a quantum critical point [29].

In this Letter, we investigate field-induced effects in high-quality single crystals of $\text{Na}_3\text{Co}_2\text{SbO}_6$ in both in-plane and out-of-plane directions, employing a variety of magnetic, thermodynamic, and theoretical methods. Our findings reveal a rich anisotropic magnetic phase diagram, highlighting field-induced magnetic phases that we characterize within an extended Kitaev-Heisenberg model with additional ring exchange interactions.

Methods. Single crystals of $\text{Na}_3\text{Co}_2\text{SbO}_6$ were grown using the flux method and were fully characterized by powder and single-crystal x-ray diffraction (SCXRD) and energy-dispersive x-ray (EDX) analysis [37]. DC magnetic-susceptibility and specific-heat measurements were carried out using a commercial superconducting quantum interference device (SQUID) magnetometer (MPMS3) and a physical property measurement system (PPMS) from Quantum Design, respectively. Quartz and quartz paddles with two quartz half-cylinders holding the crystal vertically for out-of-plane measurements were used for in-plane and out-of-plane magnetic measurements, respectively. Specific-heat measurements were conducted using the heat-pulse relaxation technique with a ^3He insert in the PPMS.

Measurements of the bulk magnetization and magnetocaloric effect (MCE) were performed using pulsed magnetic fields at the Dresden High Magnetic Field Laboratory (HLD). Here, quasiadiabatic conditions were ensured by the short duration of the magnetic-field pulse with a rise time of a few tens of ms and by pumping the sample space to pressures below 1 mbar. The magnetization was recorded with a compensated pickup-coil magnetometer in a coaxial geometry. The background of each measurement was determined by a subsequent measurement with an empty magnetometer and otherwise identical conditions [38]. The resulting pulsed-field magnetization curves were calibrated with data recorded in the MPMS. For the measurements of the MCE, the sample temperature was monitored with a RuOx-based thermometer glued directly to the sample and corrected for magnetoresistive effects using a second sensor placed nearby.

Experimental results. Figure 1(a) illustrates the $C2/m$ crystal structure of $\text{Na}_3\text{Co}_2\text{SbO}_6$. The refinement of our SCXRD data resulted in lattice parameters $a = 5.3855(3)$ Å, $b = 9.2938(3)$ Å, $c = 5.6662(2)$ Å, and $\beta = 108.6^\circ$ at room temperature. The lattice parameters indicate a tiny orthorhombic distortion, with $|\sqrt{3}a/b - 1| \lesssim 0.4\%$, consistent with previous reports [22]. The O-Co-O bond angles of the CoO_6 octahedra are $\sim 95^\circ$, indicating a significant compression along the interlayer stacking axis $\mathbf{c}^*$. Our refinement shows no detectable mixing of Co and Sb sites and the EDX analysis reveals a homogeneous elemental distribution, indicative of the high quality of our single crystals.

The temperature dependence of the DC magnetic susceptibility χ , with applied magnetic fields aligned along the in-plane directions $\mathbf{a}$ and $\mathbf{b}$, as well as the out-of-plane direction $\mathbf{c}^*$, is shown in Fig. 1(b). A sharp decrease of the magnetic susceptibility is observed below $T_N = 7$ K, indicating the onset of long-range antiferromagnetic (AFM) order.

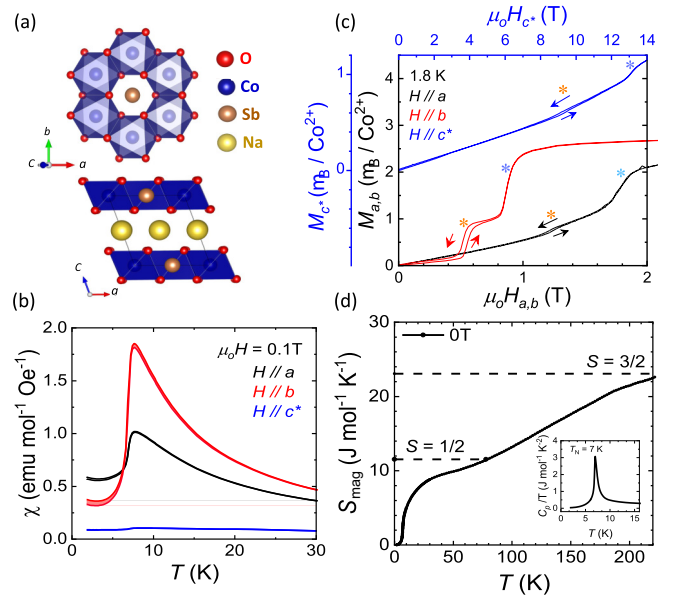


FIG. 1. (a) Room-temperature crystal structure of $\text{Na}_3\text{Co}_2\text{SbO}_6$. (b) Magnetic susceptibility χ as a function of temperature T at a field of $\mu_0 H = 0.1$ T along the crystallographic $\mathbf{a}$, $\mathbf{b}$, and $\mathbf{c}^* \perp \mathbf{a}, \mathbf{b}$ directions. The shaded areas highlight the bifurcation between field-cooled (upper curve) and zero-field-cooled (lower curve) magnetic susceptibility. (c) Isothermal magnetization M as a function of field H at $T = 1.8$ K for $\mathbf{H} \parallel \mathbf{a}$ and $\mathbf{H} \parallel \mathbf{b}$ up to 2 T and at $T = 2$ K for $\mathbf{H} \parallel \mathbf{c}^*$ up to 14 T. Asterisks indicate the field-induced transitions (field-sweep up). (d) Magnetic entropy S_{mag} as a function of temperature T at zero field. The inset displays the zero-field heat capacity C_p/T .

Here, T_N is defined as the inflection point in $\partial(\chi T)/\partial T$ according to Fisher's heat capacity definition [39] and it closely matches the Néel temperature reported previously [22]. The Na content of seven crystals from the same batch that was used for this study was checked with respect to their magnetic transition (see, e.g., Refs. [19,30]). All seven crystals order at around 7 K indicating a near-ideal Na stoichiometry. The variation of their transition temperature is less than 0.3 K, consistent with a very homogeneous Na distribution in individual crystals and, consequently, a homogeneous and near ideal Na content in the crystals that are discussed in the work at hand. The presence of magnetic order indicates the existence of non-Kitaev interactions, likely originating from the distorted octahedral environment [Co-O-Co bond angle of $\sim 92.7(4)^\circ$ and intraorbital hopping [17]. The tiny splitting between zero-field-cooled and field-cooled magnetic susceptibility below T_N indicates a weak uncompensated magnetic moment superimposed on the AFM order, consistent with previous observations [22].

Figure 1(c) shows the isothermal magnetization M as a function of field at $T = 1.8$ K for $\mathbf{H} \parallel \mathbf{a}$ and $\mathbf{H} \parallel \mathbf{b}$ up to 2 T and at $T = 2$ K for $\mathbf{H} \parallel \mathbf{c}^*$ up to 14 T. Our $M(H)$ data reveal multiple distinct field-induced transitions, some of them showing strong hysteresis effects. Interestingly, field-induced transitions are also observed in the out-of-plane $\mathbf{c}^*$ direction, which was overlooked in previous studies [21,22,30], highlighting the complex magnetic anisotropy in $\text{Na}_3\text{Co}_2\text{SbO}_6$. The observed critical field values vary by an

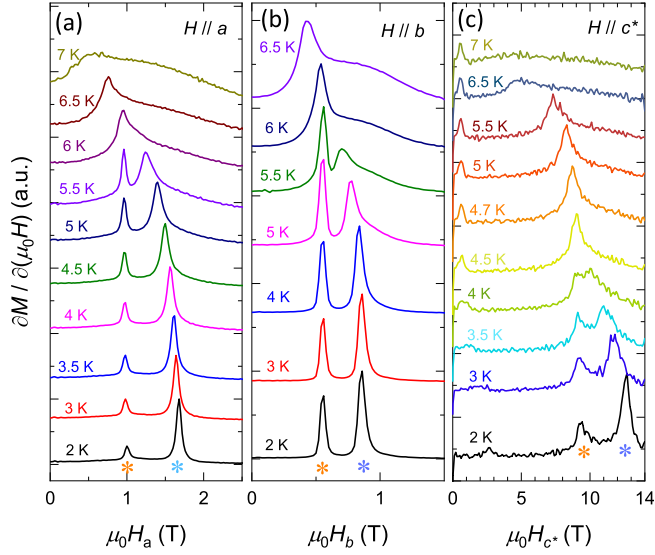


FIG. 2. (a) Magnetic susceptibility $\partial M / \partial(\mu_0 H)$ for various fixed temperatures as a function of field $\mathbf{H} \parallel \mathbf{a}$ (field-sweep up) from SQUID magnetometry up to 2.5 T. Asterisks indicate the field-induced transitions. (b) Same as (a), but for $\mathbf{H} \parallel \mathbf{b}$ up to 1.5 T. (c) Same as (a), but for $\mathbf{H} \parallel \mathbf{c}^*$ from vibrating sample magnetometry in a PPMS up to 14 T.

order of magnitude depending on the direction, with the lowest field values found along the in-plane easy axis $\mathbf{b}$ and the highest values along the out-of-plane direction $\mathbf{c}^*$. The critical field values of the field-induced transitions for increasing magnetic fields (field-up sweeps) are obtained from the peak of the anomaly in the derivative curves $\partial M / \partial(\mu_0 H)$ at different temperatures, shown in Fig. 2. For fields along the in-plane direction $\mathbf{a}$ [Fig. 2(a)] and $T = 2$ K, we observe two field-induced transitions at $\mu_0 H_{a1} = 0.99(50)$ T and $\mu_0 H_{a2} = 1.68(12)$ T. For fields along the in-plane direction $\mathbf{b}$ [Fig. 2(b)] and $T = 2$ K, we observe two transitions at $\mu_0 H_{b1} = 0.56(8)$ T and $\mu_0 H_{b2} = 0.86(5)$ T, with the latter marking the complete suppression of the AFM order. The specific shape of $\partial M / \partial(\mu_0 H)$ near H_{b2} for $T = 5$ K might hint at short-range order in this region, in line with our specific-heat studies presented in the Supplemental Material (SM) [37]. Our results for the two in-plane directions are consistent with the literature [22]. Note that the critical fields along $\mathbf{a}$ exhibit large uncertainties due to a possible misorientation (in this work $\lesssim 10^\circ$) due to steep changes in the critical fields close to $\mathbf{a}$, in contrast to the $\mathbf{b}$ direction (see the Supplemental Material [37]).

For fields along the out-of-plane direction $\mathbf{c}^*$, the critical field strengths are significantly larger. For $T = 2$ K, we observe two field-induced transitions at $\mu_0 H_{c1} = 9.4(4)$ T and $\mu_0 H_{c2} = 12.6(8)$ T [see Fig. 2(c)]. The anomaly in the field range $0 \leq \mu_0 H \leq 2.6$ T probably arises from short-range ferromagnetic chains formed above T_N [22] as this anomaly persists in the paramagnetic phase [37]. Note that all transitions were consistently reproduced across different single-crystal samples, with slight variations in the critical field strengths ($\lesssim 0.4$ T and $\lesssim 2$ T for in and out of plane, respectively), likely due to minor differences in stoichiometry,

disorder, or orientation. For all three directions, the $M(H)$ curves at 2 K exhibit clear hysteresis at the lowest-field transition, indicating a first-order nature, which is further confirmed by our MCE measurements discussed below.

Overall, the isothermal magnetization reveals pronounced magnetic anisotropy. From the quasaturated magnetizations, we estimate the anisotropic g factors as $g_a = 6.2 \pm 0.3$, $g_b = 6.4 \pm 0.3$, and $g_{c^*} = 3.6 \pm 0.4$; see SM [37] for details. These values contrast with those of an undistorted octahedron, which is expected to exhibit an isotropic $g_{\text{iso}} = 4.33$ for a $j_{\text{eff}} = 1/2$ doublet ground state [40]. In $\text{Na}_3\text{Co}_2\text{SbO}_6$, the distortion of the CoO_6 octahedra induces an admixture of $m = \pm 1/2$ components from the $j_{\text{eff}} = 3/2$ and $j_{\text{eff}} = 5/2$ multiplets with the $j_{\text{eff}} = 1/2$ state, giving rise to off-diagonal Γ' interactions [41]. The significantly larger values of g_a and g_b (perpendicular to the trigonal field direction) compared to g_{c^*} suggest that the CoO_6 octahedra are compressed along the trigonal field direction, in agreement with the SCXRD results. Applying biaxial compressive strain in the ab plane could thus be a viable approach to achieve the ideal, undistorted octahedral environment in $\text{Na}_3\text{Co}_2\text{SbO}_6$, similar to $\text{Cu}_3\text{Co}_2\text{SbO}_6$ [42].

The inset of Fig. 1(d) shows the specific heat C_p/T at zero field as a function of temperature. A clear peak at $T_N = 7$ K indicates the onset of long-range magnetic order. No additional sharp anomalies were detected in our specific-heat measurements up to 220 K (see SM [37]). The magnetic contribution to the zero-field specific heat was extracted by subtracting the phonon contribution, obtained from the non-magnetic analog compound $\text{Na}_3\text{Zn}_2\text{SbO}_6$. The corresponding zero-field magnetic entropy $S_{\text{mag}} = \int dT C_{\text{mag}}/T$ is shown in Fig. 1(d). For temperatures $T \lesssim 50$ –80 K, the entropy reaches a plateau at around $S_{\text{mag}}^{\text{theory}} = 2R \ln 2 = 11.5$ J/mol/K, consistent with the expectation for a system containing two magnetic Co^{2+} ions with $j_{\text{eff}} = 1/2$. The accumulated entropy up to $T_N = 7$ K represents only $\sim 15\%$ of $S_{\text{mag}}^{\text{theory}}$, indicating frustrated quasi-2D short-range spin correlations reaching up to ~ 50 K. The additional magnetic entropy release above ~ 80 K arises from excitations to $j_{\text{eff}} = 3/2$ states, consistent with the observation of an additional mode in Raman spectroscopy [43]. Our modified Curie-Weiss fit of the magnetic susceptibility for $\mathbf{H} \parallel \mathbf{c}^*$ supports this conclusion and indicates an excitation gap of $\Delta E_{\frac{1}{2} \rightarrow \frac{3}{2}} = 247(3)$ K (see SM [37]).

Our MCE measurements under quasiadiabatic conditions in pulsed magnetic fields, shown in Fig. 3 for both field sweep-up and sweep-down, provide additional insights into the nature of the field-induced magnetic phases in $\text{Na}_3\text{Co}_2\text{SbO}_6$. For these studies, a different set of single crystals was used, resulting in slightly different critical field values. In general, the critical field values are determined from the minima in $\partial T_S / \partial(\mu_0 H)$, where T_S refers to the sample temperature. This definition is consistent with an increase in spin disorder near the phase transition, which forces a drop in thermal entropy to compensate. For a first-order transition involving latent heat, however, the quasiadiabatic condition may be violated, resulting in maxima in $\partial T_S / \partial(\mu_0 H)$ due to an abrupt increase in the sample temperature. For the a direction, Fig. 3(a), during the field-up sweep, the sample temperature increases monotonically until the critical field $\mu_0 H_{a1} \sim 1.5$ T, indicating a decrease in magnetic entropy. At the critical field

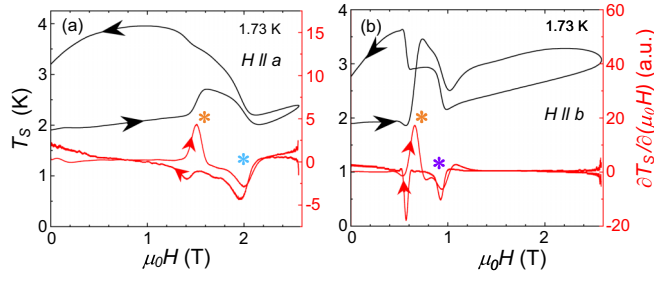


FIG. 3. (a) Sample temperature T_s (black) and its derivative $\partial T_s / \partial (\mu_0 H)$ (red) as a function of field $\mathbf{H} \parallel \mathbf{a}$ from low-temperature MCE measurements in pulsed fields under quasiadiabatic conditions. Arrows indicate the field-up and field-down sweeps, while asterisks mark the field-induced transitions. (b) Same as (a), but for $\mathbf{H} \parallel \mathbf{b}$.

H_{a1} , a sudden increase in the sample temperature during the field-up sweep marks the release of latent heat. Conversely, the field-down sweep exhibits a further temperature increase below H_{a1} , leading to a negative slope $\partial T_s / \partial (\mu_0 H) < 0$, confirming the first-order nature of the phase transition.

The strong irreversibility between the sweep-up and sweep-down processes may be attributed to AFM domain dynamics and the complex field evolution of the double- $\mathbf{q}$ spin structure [22], with a loss mechanism driven by spin fluctuations. A local minimum in T_s marks the critical field H_{a2} . This behavior is consistently observed in both field-up and field-down sweeps, suggesting the second-order nature of this transition. Above H_{a2} , the steady increase in T_s reflects the opening of a spin-excitation gap in the field-polarized state, compensating for the drop in spin entropy. This is in line with

our specific-heat measurements for fields above H_{a2} (see SM [37]). A similar behavior has also been observed for the b direction, where the field-induced transitions at the two critical fields are marked by a sudden increase and a local minimum in the sample temperature, respectively, corresponding to first-order (H_{b1}) and second-order (H_{b2}) phase transitions. Contrary to the a direction, a strong hysteresis is observed above H_{b2} in the field-polarized state, suggesting some irreversibility arising from mechanisms beyond simple magnetic-domain effects. For the c^* axis, the critical field H_{c2} is identified for field-up sweeps in our MCE study; see SM [37].

The anisotropic field and temperature evolution of the magnetic ground state of $\text{Na}_3\text{Co}_2\text{SbO}_6$ as determined through magnetic and specific heat measurements is summarized in Figs. 4(a)–4(c) for the main crystallographic directions $\mathbf{a}$, $\mathbf{b}$, and $\mathbf{c}^*$. Along the b direction, in the field range $0.7 \leq \mu_0 H \leq 0.85$ T, just after the suppression of magnetic long-range order, an additional anomaly emerges, possibly due to short-range magnetic order or a small domain rotated by 60° , indicated by dark red stars in Fig. 4(b). Increasing the magnetic field beyond the suppression of AFM order leads to the opening of a spin-excitation gap, as revealed by our specific-heat measurements; see insets of Figs. 4(a) and 4(b) and the SM [37]. We note that it is beyond the scope of this work to speculate on the existence of a possible gapless regime between the full suppression of long-range AFM order and the opening of the spin gap. However, if such a regime exists, its width would not exceed $\Delta \mu_0 H \sim 0.5$ T.

Modeling. In order to qualitatively characterize the experimentally observed field-induced phases theoretically, we use classical Monte Carlo simulations [32,33] to study an ex-

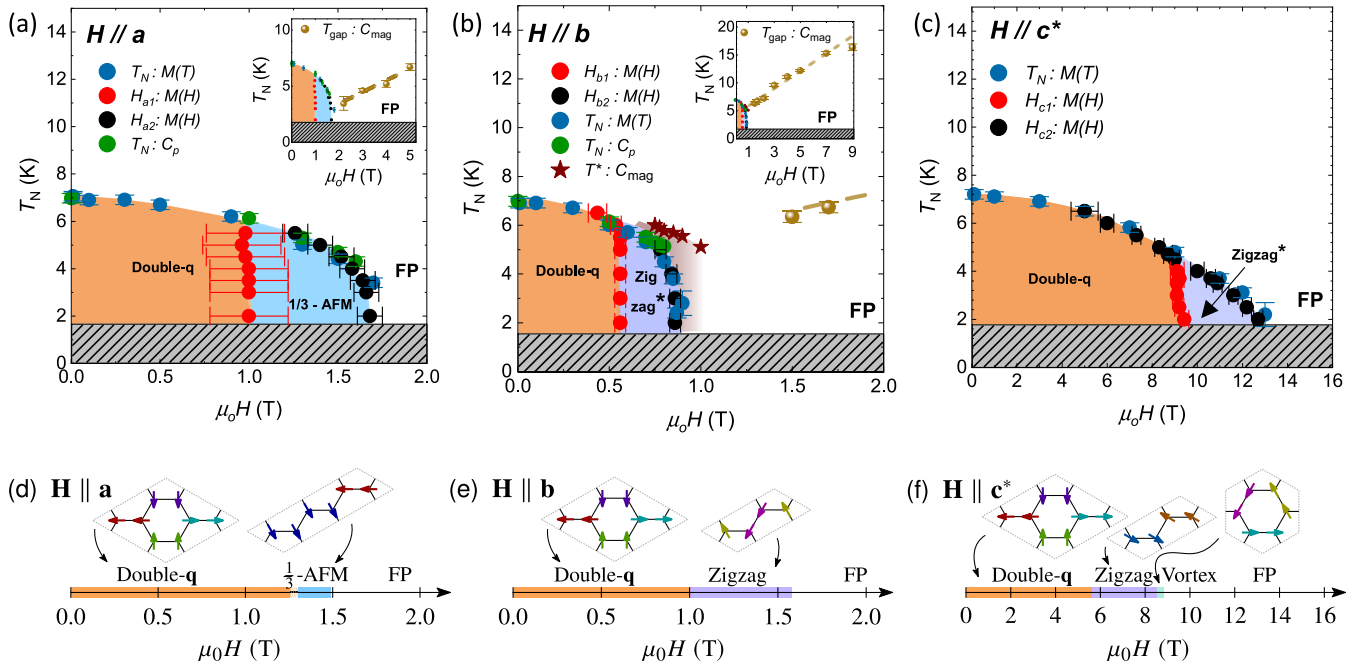


FIG. 4. Top: $\text{Na}_3\text{Co}_2\text{SbO}_6$ experimental magnetic phase diagram for (a) $\mathbf{H} \parallel \mathbf{a}$, (b) $\mathbf{H} \parallel \mathbf{b}$, and (c) $\mathbf{H} \parallel \mathbf{c}^*$. Note that the phase boundaries correspond to the critical fields observed for field-up sweeps. The field-induced phases predicted by theory are marked with an asterisk, while those without an asterisk are based on previous neutron diffraction experiments [22]. Insets illustrate the spin-excitation gap in the field-polarized (FP) state. Bottom: ground-state phase diagram of extended Kitaev-Heisenberg model for (d) $\mathbf{H} \parallel \mathbf{a}$, (e) $\mathbf{H} \parallel \mathbf{b}$, and (f) $\mathbf{H} \parallel \mathbf{c}^*$. Insets indicate spin directions projected onto the ab plane in the different phases.

tended Kitaev-Heisenberg model, defined by the Hamiltonian

$$\begin{aligned} \mathcal{H} = & \sum_{\langle ij \rangle_\gamma} [J \mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \\ & + \Gamma' (S_i^\gamma S_j^\alpha + S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\beta + S_i^\beta S_j^\gamma)] + \mathcal{H}_\square^C \\ & - \mu_0 \mathbf{H} \cdot \sum_i \mu_B g \mathbf{S}_i, \end{aligned} \quad (1)$$

with nearest-neighbor bilinear exchange couplings J , K , Γ , and Γ' , a nonbilinear ring exchange $\mathcal{H}_\square^C$, a Zeeman coupling of the effective moment $\mathbf{M} = \sum_i \mu_B g \mathbf{S}_i$ to the magnetic field $\mu_0 \mathbf{H}$, and a diagonal g tensor $g = \text{diag}(g_a, g_b, g_{c^*})$, with g_{a,b,c^*} taken from the high-field magnetization measurements discussed above. Ring exchange interactions have recently been argued to be of relevance in the sister compound $\text{Na}_2\text{Co}_2\text{TeO}_6$ [31–34]. Here, we assume a form consistent with the C_2^* of the monoclinic lattice [22], $\mathcal{H}_\square^C = J_\square \sum_{(ijklmn)} [(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_j \cdot \mathbf{S}_k)(\mathbf{S}_m \cdot \mathbf{S}_n) + (\mathbf{S}_i \cdot \mathbf{S}_k)(\mathbf{S}_j \cdot \mathbf{S}_m)(\mathbf{S}_l \cdot \mathbf{S}_n) + (ikm) \leftrightarrow (jnl)]$, with the summation over elemental plaquettes involving sites $(ijklmn)$ in counterclockwise order, and $(ikm) \leftrightarrow (jnl)$ denotes terms arising from the first two terms by an exchange of lattice indices, corresponding to symmetrization with respect to the C_2^* symmetry. Details of the simulations are given in the SM [37]. We choose $(J, K, \Gamma, \Gamma', J_\square) = A(-1/9, -2/3, 3, -4/9, -3/20)$, where $A = 0.5$ meV corresponds to the overall energy scale. These parameters have been chosen to recover (1) the zero-field double- $\mathbf{q}$ ground state found in neutron diffraction experiments [22] and (2) the sizable in-plane vs out-of-plane magnetic anisotropy manifested in Figs. 4(a)–4(c). Remarkably, as illustrated in the ground-state phase diagrams shown in Figs. 4(d)–4(f), the model constructed this way automatically reproduces several further features of $\text{Na}_3\text{Co}_2\text{SbO}_6$ found in our measurements and previous literature results, namely (3) the presence of field-induced intermediate ordered phases for all three field directions and (4) an evolution of the spin structure factor with Bragg peaks at momenta $\mathbf{q} = \mathbf{M}$ (double- $\mathbf{q}$ phase) for small fields, shifting to $\mathbf{q} = (2/3)\mathbf{M}$ for fields $\mathbf{H} \parallel \mathbf{a}$ in the phase below the high-field transition (1/3-AFM phase), in agreement with the neutron diffraction results [22]. The agreement between theory and experiment allows us to predict the nature of the field-induced phases that have previously not been characterized in the literature. In particular, our modeling suggests that the field-induced intermediate ordered phases observed for $\mathbf{H} \parallel \mathbf{b}$ and $\mathbf{H} \parallel \mathbf{c}^*$ realize canted zigzag order, in contrast to the field-induced 1/3-AFM order observed for $\mathbf{H} \parallel \mathbf{a}$. The conclusion that the field-induced orders for $\mathbf{H} \parallel \mathbf{a}$ and $\mathbf{H} \parallel \mathbf{b}$ are not adiabatically connected is supported by magnetization measurements as a function of the in-plane

field angle, presented in the SM [37]. The new phases predicted from the modeling are marked with an asterisk in Figs. 4(a)–4(c). Corresponding real-space spin configurations are sketched in the insets of Figs. 4(d)–4(f) and further characterization is presented in the SM [37].

Conclusions. Our study on bulk single crystals of $\text{Na}_3\text{Co}_2\text{SbO}_6$ reveals an AFM order below T_N with strong in-plane versus out-of-plane anisotropy. Our results establish a $j_{\text{eff}} = \frac{1}{2}$ ground state that is well separated from the excited $j_{\text{eff}} = \frac{3}{2}$ state at low temperatures. The anisotropic magnetic phase diagram was constructed from temperature- and field-dependent specific-heat and magnetization measurements along all three crystallographic axes, revealing multiple field-induced phases. The nature of the transitions has been determined through quasiadiabatic magnetocaloric effect studies. Using classical Monte Carlo simulations within an extended K - J - Γ - Γ' Kitaev-Heisenberg model that incorporates C_2 ring exchange interactions, we have characterized the experimentally observed field-induced phases. Our results call for neutron diffraction studies to validate the field-induced canted zigzag orders predicted for fields along the b and c^* directions. Measuring the excitation spectrum using inelastic neutron scattering in the field-polarized phase could help to determine the most suitable effective spin model for $\text{Na}_3\text{Co}_2\text{SbO}_6$, similar to recent work on $\text{BaCo}_2(\text{AsO}_4)_2$ [44].

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Data availability. The data that support the findings of this Letter are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

- [1] L. Savary and L. Balents, Quantum spin liquids: A review, *Rep. Prog. Phys.* **80**, 016502 (2017).
- [2] V. Lohani, C. Hickey, J. Masell, and A. Rosch, Quantum skyrmions in frustrated ferromagnets, *Phys. Rev. X* **9**, 041063 (2019).
- [3] L. Janssen, E. C. Andrade, and M. Vojta, Honeycomb-lattice Heisenberg-Kitaev model in a magnetic field: Spin canting,

metamagnetism, and vortex crystals, *Phys. Rev. Lett.* **117**, 277202 (2016).

- [4] X. Li, C. Xu, B. Liu, X. Li, L. Bellaiche, and H. Xiang, Realistic spin model for multiferroic NiI_2 , *Phys. Rev. Lett.* **131**, 036701 (2023).
- [5] C. Kim, O. Vilella, Y. Lee, P. Park, Y. An, W. Cho, M. B. Stone, A. I. Kolesnikov, Y. Hao, S. Asai, S. Itoh, T. Masuda,

- S. Matin, Y. Kim, S. J. Kim, M. Mourigal, and J. G. Park, Kitaev interaction and proximate higher-order skyrmion crystal in the triangular lattice van der Waals antiferromagnet NiI_2 , *npj Quantum Mater.* **11**, 20 (2026).
- [6] S. Guan, W. Duan, and X. Zou, Controlled separation of skyrmions and antiskyrmions by Kitaev interaction, *Nano Lett.* **24**, 15076 (2024).
- [7] X. Jin, M. Geng, F. Orlandi, D. Khalyavin, P. Manuel, Y. Liu, and Y. Li, Robust triple-q magnetic order with trainable spin vorticity in $\text{Na}_2\text{Co}_2\text{TeO}_6$, *Phys. Rev. Lett.* **135**, 136701 (2025).
- [8] 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).
- [9] R. Sano, Y. Kato, and Y. Motome, Kitaev-Heisenberg Hamiltonian for high-spin d^7 Mott insulators, *Phys. Rev. B* **97**, 014408 (2018).
- [10] H. Liu, J. Chaloupka, and G. Khaliullin, Kitaev spin liquid in $3d$ transition metal compounds, *Phys. Rev. Lett.* **125**, 047201 (2020).
- [11] C. Kim, H.-S. Kim, and J.-G. Park, Spin-orbital entangled state and realization of Kitaev physics in $3d$ cobalt compounds: A progress report, *J. Phys.: Condens. Matter* **34**, 023001 (2022).
- [12] I. Rousochatzakis, N. B. Perkins, Q. Luo, and H.-Y. Kee, Beyond Kitaev physics in strong spin-orbit coupled magnets, *Rep. Prog. Phys.* **87**, 026502 (2024).
- [13] S. M. Winter, Magnetic couplings in edge-sharing high-spin d^7 compounds, *J. Phys. Mater.* **5**, 045003 (2022).
- [14] L. Janssen and M. Vojta, Heisenberg-Kitaev physics in magnetic fields, *J. Phys.: Condens. Matter* **31**, 423002 (2019).
- [15] 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).
- [16] B. S. Mou, X. Zhang, L. Xiang, Y. Xu, R. Zhong, R. J. Cava, H. Zhou, Z. Jiang, D. Smirnov, N. Drichko, and S. M. Winter, Comparative Raman scattering study of crystal field excitations in Co-based quantum magnets, *Phys. Rev. Mater.* **8**, 084408 (2024).
- [17] X. Liu and H.-Y. Kee, Non-Kitaev versus Kitaev honeycomb cobaltates, *Phys. Rev. B* **107**, 054420 (2023).
- [18] M. Songvilay, J. Robert, S. Petit, J. A. Rodriguez-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 $\text{Na}_3\text{Co}_2\text{SbO}_6$ and $\text{Na}_2\text{Co}_2\text{TeO}_6$, *Phys. Rev. B* **102**, 224429 (2020).
- [19] 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).
- [20] M. I. Stratan, I. L. Shukaev, T. M. Vasilchikova, A. N. Vasiliev, A. N. Korshunov, A. I. Kurbakov, V. B. Nalbandyan, and E. A. Zvereva, Synthesis, structure and magnetic properties of honeycomb-layered $\text{Li}_3\text{Co}_2\text{SbO}_6$ with new data on its sodium precursor, $\text{Na}_3\text{Co}_2\text{SbO}_6$, *New J. Chem.* **43**, 13545 (2019).
- [21] 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).
- [22] 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).
- [23] C. Kim, J. Jeong, G. Lin, P. Park, T. Masuda, S. Asai, S. Itoh, H.-S. Kim, H. Zhou, J. Ma, and J.-G. Park, Antiferromagnetic Kitaev interaction in $J_{\text{eff}} = 1/2$ cobalt honeycomb materials $\text{Na}_3\text{Co}_2\text{SbO}_6$ and $\text{Na}_2\text{Co}_2\text{TeO}_6$, *J. Phys.: Condens. Matter* **34**, 045802 (2022).
- [24] 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 $\text{Na}_3\text{Co}_2\text{SbO}_6$ and $\text{Na}_2\text{Co}_2\text{TeO}_6$, *Phys. Rev. B* **106**, 014413 (2022).
- [25] E. Vavilova, T. Vasilchikova, A. Vasiliev, D. Mikhailova, V. Nalbandyan, E. Zvereva, and S. V. Streltsov, Magnetic phase diagram and possible Kitaev-like behavior of the honeycomb-lattice antimonate $\text{Na}_3\text{Co}_2\text{SbO}_6$, *Phys. Rev. B* **107**, 054411 (2023).
- [26] 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).
- [27] 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).
- [28] 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).
- [29] 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).
- [30] 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).
- [31] W. G. F. Krüger, W. Chen, X. Jin, Y. Li, and L. Janssen, Triple-q order in $\text{Na}_2\text{Co}_2\text{TeO}_6$ from proximity to hidden-SU(2)-symmetric point, *Phys. Rev. Lett.* **131**, 146702 (2023).
- [32] N. Francini and L. Janssen, Spin vestigial orders in extended Heisenberg-Kitaev models near hidden SU(2) points: Application to $\text{Na}_2\text{Co}_2\text{TeO}_6$, *Phys. Rev. B* **109**, 075104 (2024).
- [33] N. Francini and L. Janssen, Ferrimagnetism from triple-q order in $\text{Na}_2\text{Co}_2\text{TeO}_6$, *Phys. Rev. B* **110**, 235118 (2024).
- [34] N. Francini, P. M. Cónsoli, and L. Janssen, Ferrimagnetism from quantum fluctuations in Kitaev materials, *Phys. Rev. B* **112**, 155103 (2025).
- [35] P. Park, W. Cho, C. Kim, Y. An, Y.-G. Kang, M. Avdeev, R. Sibille, K. Iida, R. Kajimoto, K. H. Lee, W. Ju, E.-J. Cho, H.-J. Noh, M. J. Han, S.-S. Zhang, C. D. Batista, and J.-G. Park, Tetrahedral triple-Q magnetic ordering and large spontaneous Hall conductivity in the metallic triangular antiferromagnet $\text{Co}_{1/3}\text{TaS}_2$, *Nat. Commun.* **14**, 8346 (2023).
- [36] N. D. Andriushin, J. Grumbach, A. A. Kulbakov, Y. V. Tymoshenko, Y. A. Onykienko, R. Firouzmandi, E. Cheng,

- S. Granovsky, Y. Skourski, J. Ollivier, H. C. Walker, V. Kocsis, B. Büchner, B. Keimer, M. Doerr, D. S. Inosov, and D. C. Peets, Anomalous quasielastic scattering contribution in the centrosymmetric multi- $\mathbf{q}$ helimagnet SrFeO_3 , *Phys. Rev. X* **15**, 011038 (2025).
- [37] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/r454-vd8b> for additional information on (i) the crystal growth and the structural analysis of our single crystals, (ii) DC magnetization measurements in static and pulsed fields, along with further analysis, (iii) specific-heat measurements of $\text{Na}_3\text{Co}_2\text{SbO}_6$ and its nonmagnetic analog compound $\text{Na}_3\text{Zn}_2\text{SbO}_6$, and (iv) details of the Monte Carlo simulations, along with further characterization of the field-induced phases, which also includes Refs. [3,29,33,47–52].
- [38] Y. Skourski, M. D. Kuz'min, K. P. Skokov, A. V. Andreev, and J. Wosnitzer, High-field magnetization of $\text{Ho}_2\text{Fe}_{17}$, *Phys. Rev. B* **83**, 214420 (2011).
- [39] M. E. Fisher, Relation between the specific heat and susceptibility of an antiferromagnet, *Philos. Mag.* **7**, 1731 (1962).
- [40] A. Abragam and B. Bleaney, *Electron Paramagnetic Resonance of Transition Ions* (Oxford University Press, Oxford, 2012).
- [41] J. G. Rau and H.-Y. Kee, Trigonal distortion in the honeycomb iridates: Proximity of zigzag and spiral phases in Na_2IrO_3 , [arXiv:1408.4811](https://arxiv.org/abs/1408.4811).
- [42] G.-H. Kim, M. Park, S. Samanta, U. Choi, B. Kang, U. Seo, G. Ji, S. Noh, D.-Y. Cho, J.-W. Yoo, J. M. Ok, H.-S. Kim, and C. Sohn, Suppression of antiferromagnetic order by strain-enhanced frustration in honeycomb cobaltate, *Sci. Adv.* **10**, eadn8694 (2024).
- [43] Y. S. Ponomov, E. V. Komleva, E. A. Pankrushina, D. Mikhailova, and S. V. Streltsov, Raman spectroscopy of $\text{Na}_3\text{Co}_2\text{SbO}_6$, *JETP Lett.* **119**, 518 (2024).
- [44] P. A. Maksimov, S. Jiang, L. P. Regnault, and A. L. Chernyshev, Strong Kitaev interaction in $\text{BaCo}_2(\text{AsO}_4)_2$, *Phys. Rev. Lett.* **135**, 066703 (2025).
- [45] K. Momma and F. Izumi, *VESTA3* for three-dimensional visualization of crystal, volumetric and morphology data, *J. Appl. Cryst.* **44**, 1272 (2011).
- [46] <http://www.nhr-verein.de/en/our-partners>
- [47] CrysAlis PRO, Agilent Technologies Ltd, Yarnton, Oxfordshire, England, 2014.
- [48] V. Petříček, L. Palatinus, J. Plášil, and M. Dušek, Jana2020 – a new version of the crystallographic computing system Jana, *Z. Kristallogr. Cryst. Mater.* **238**, 271 (2023).
- [49] S. Zhang, S. Lee, E. Brosha, Q. Huang, H. Zhou, V. S. Zapf, and M. Lee, Out-of-plane magnetic phase diagram of the Kitaev quantum magnet $\text{Na}_2\text{Co}_2\text{TeO}_6$, *Phys. Rev. B* **110**, 144431 (2024).
- [50] T. Besara, M. S. Lundberg, J. Sun, D. Ramirez, L. Dong, J. B. Whalen, R. Vasquez, F. Herrera, J. R. Allen, M. W. Davidson, and T. Siegrist, Single crystal synthesis and magnetism of the BaLn_2O_4 family (Ln = lanthanide), *Prog. Solid State Chem.* **42**, 23 (2014).
- [51] A. Tari, *The Specific Heat of Matter at Low Temperatures* (Imperial College Press, World Scientific, Singapore, 2003).
- [52] L. Janssen, E. C. Andrade, and M. Vojta, Magnetization processes of zigzag states on the honeycomb lattice: Identifying spin models for $\alpha\text{-RuCl}_3$ and Na_2IrO_3 , *Phys. Rev. B* **96**, 064430 (2017).