

Trigonal distortion in the Kitaev candidate honeycomb magnet $\text{BaCo}_2(\text{AsO}_4)_2$

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We conducted x-ray absorption (XAS) and magnetic circular dichroism (XMCD) measurements at the Co $L_{2,3}$ edges on single crystals of the Kitaev candidate honeycomb lattice compound $\text{BaCo}_2(\text{AsO}_4)_2$. The measurements employed the inverse partial fluorescence yield technique, which is ideal for acquiring reliable x-ray absorption spectra from highly insulating samples, enabling precise quantitative analysis. Our experimental results revealed a significant linear dichroic signal, indicating strong trigonal distortion in the CoO_6 octahedra in $\text{BaCo}_2(\text{AsO}_4)_2$. We performed a detailed analysis of the experimental XAS and XMCD spectra using a full-multiplet configuration-interaction cluster model. This analysis unveiled that the t_{2g} hole density is predominantly localized in the a_{1g} orbital. Through XMCD sum rules and theoretical calculations, we quantified both the spin and orbital magnetic moments. Our study demonstrates that the local electronic structure of the CoO_6 octahedra displays an effective trigonal distortion of approximately -0.114 eV. This distortion is larger than the Co $3d$ spin-orbit coupling constant, emphasizing the crucial impact of local structural distortions on the electronic and magnetic properties of $\text{BaCo}_2(\text{AsO}_4)_2$.

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I. INTRODUCTION

Novel and exotic quantum phases of matter display a plethora of physics that conventional mean field theories may not capture. One of such states is the quantum spin liquid (QSL) ground state, which does not exhibit long-range magnetic order but is characterized by topological order with fractionalized excitations and long-range entanglement, displaying highly correlated spins that fluctuate even at absolute zero temperature [1]. Such systems were initially explored in geometrically frustrated magnetic structures with $S = 1/2$, but recently the search for the materialization of the QSL ground state became tremendously intensified with the theoretical implementation of the exactly solvable Kitaev model [2] for $J_{\text{eff}} = 1/2$ pseudospin systems with the honeycomb lattice structure [3]. In this model, neighboring spins are coupled in a highly anisotropic manner with bond-dependent interactions originating from the spin-orbit coupling, which give rise to localized and itinerant Majorana fermions.

Initially, most studies were focused on compounds containing $4d$ and $5d$ transition metal ions (i.e., Ru^{3+} , Rh^{4+} , and Ir^{4+}) in edge-sharing octahedral geometry [4–6]. However, it was found that the real-world honeycomb materials always feature sizable non-Kitaev interactions [7] that lead to magnetic order at low temperatures. Yet the expectation remains that such materials may be tuned into the QSL phase via external parameters such as pressure, uniaxial strain, or applied magnetic field. In $\alpha\text{-RuCl}_3$, for example, the zig-zag antiferromagnetic order which appears at $T_N \approx 7\text{--}13$ K could be suppressed by the application of an in-plane magnetic field of ≈ 8 T [8,9]. This field-induced phase was quickly characterized as the Kitaev QSL phase [10]. Nevertheless, this assessment is currently still subject of intense debate [6].

Very recently, it has been proposed that honeycomb lattice based compounds with Co^{2+} ions in octahedral coordination could be suitable candidates for the realization of a QSL ground state [11,12]. In the presence of the octahedral crystal field, the high-spin $3d^7$: $t_{2g}^5 e_g^2$ ($S = 3/2$, $L_{\text{eff}} = 1$) configuration also possesses the effective $J_{\text{eff}} = 1/2$ ground state due to the spin-orbit coupling (SOC), very similar to the $4d^5$ and $5d^5$ Kitaev candidate materials. Moreover, the presence of the e_g electrons in the ground state configuration leads to more exchange channels which should further suppress the Heisenberg and off-diagonal exchange interactions due to their opposite sign [13]. Thus, Co^{2+} honeycomb materials seem to provide improved conditions for the realization of

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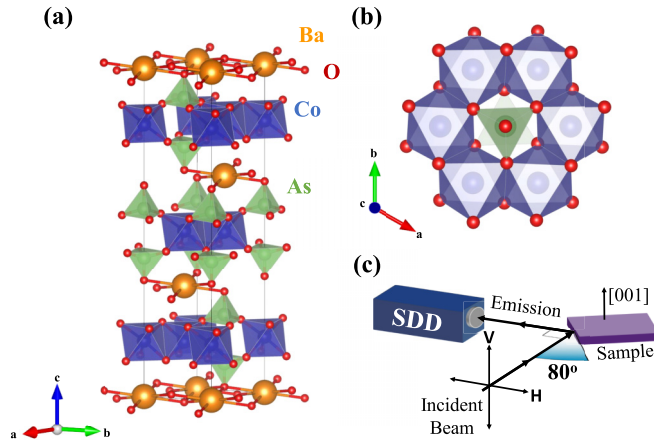


FIG. 1. (a) Crystal structure of $\text{BaCo}_2(\text{AsO}_4)_2$ displaying layered arrangement of CoO_6 octahedra along the c axis. (b) The ab plane containing honeycomb lattice of edge-sharing CoO_6 octahedra with AsO_4 tetrahedra at the center. (c) The experimental geometry of the incident beam, sample, and SDD detector.

the Kitaev QSL ground state. However, details of the actual crystal structure of the material may matter since the presence of effective crystal fields of lower than octahedral symmetry could cause serious deviations from the ideal $J_{\text{eff}} = 1/2$ state [14,15]. This is the focus of our study on the candidate material $\text{BaCo}_2(\text{AsO}_4)_2$ in which we determine the local electronic and magnetic properties of the Co^{2+} ions using linearly polarized x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) at the $L_{2,3}$ edges.

$\text{BaCo}_2(\text{AsO}_4)_2$ shows several intriguing properties, most notably, the suppression of long-range magnetic order by in-plane magnetic field of ≈ 0.5 T [16]. It crystallizes in the hexagonal $R\bar{3}$ space group, consisting of layers of edge-sharing Co^{2+} octahedra which are sandwiched in between AsO_4 tetrahedra along the c axis as depicted in Fig. 1(a). The edge-sharing CoO_6 octahedra forming the honeycomb lattice in the ab plane is shown in Fig. 1(b). In this structure, the Co^{2+} ions have a site symmetry C_3 . The material orders antiferromagnetically at the temperature $T_N \approx 5.4$ K. The suppression of magnetic order by a small magnetic field suggests a possible field-induced Kitaev QSL ground state [16]. A finite residual linear term of thermal conductivity observed in this field regime has been attributed to mobile fractionalized spinon excitation [17].

On the other hand, magnon excitations found in time domain terahertz spectroscopy measurements [18] have indicated that the field-induced state in the field regime of 0.5 T is not compatible with a Kitaev QSL. Furthermore, a recent study consisting of magnetometry, calorimetry, high-resolution capacitive dilatometry, and single crystal neutron diffraction experiments reported that a magnetic Bragg peak persists in the proposed QSL phase with applied in-plane magnetic fields [19]. However, the exact microscopic model of this material, the size and even the sign of the Kitaev term remain vividly debated [20–23]. In fact, one would readily expect that a trigonal distortion in these systems could modify the pseudospin wave function, affecting the non-Kitaev terms [13,15,24] and thus strongly influencing the J/K ratio.

Hence it is vitally important to determine the magnitude of the trigonal distortion since this gives an estimate of the energy scale required for tuning the system into the QSL regime.

In this context, XAS and XMCD at the $\text{Co } L_{2,3}$ edges combined with full-multiplet configuration-interaction cluster calculations are powerful methods to determine the ground-state quantum numbers of the Co ions as well as the relevant energy parameters like effective crystal field, SOC, and trigonal distortion [25–30].

Of particular interest for Co^{2+} ions in octahedral coordination is that XAS and XMCD are very sensitive to which of the lowest spin-orbit split $J_{\text{eff}} = 1/2$, $3/2$, and $5/2$ states is occupied. This is illustrated in Fig. 2: theoretical calculations for the dipole-allowed $2p^6 3d^7 \rightarrow 2p^5 3d^8$ transitions show that each of the starting J_{eff} states produces its own specific XAS spectrum. In particular, the $J_{\text{eff}} = 5/2$ sextet is further split by the cubic crystal field into a quartet and a doublet.

The splitting between the different J_{eff} states can be very small and is definitely smaller than the experimental photon energy resolution and the inverse lifetime of the $2p$ core hole. Yet which spectrum will be detected depends solely on which J_{eff} states are occupied. This occupation will change with temperature, leading to a nontrivial behavior of the magnetic susceptibility [30] and/or due to mixing by lower-than-octahedral crystal fields. It is interesting to note that the specificity for the J_{eff} state is not only in the linear or magnetic circular polarization dependence but already in the isotropic part of the spectrum. This makes this XAS technique particularly suitable also for polycrystalline materials.

II. METHODS

The single crystal growth and basic characterization of $\text{BaCo}_2(\text{AsO}_4)_2$ samples used in this study can be found in Ref. [19]. The XAS and XMCD measurements were performed at the BL29 of Boreas beamline of the ALBA synchrotron radiation facility in Barcelona [31]. A dark pink single crystal was mounted on an aluminum sample holder such that the Poynting vector and the linear horizontal (LH) polarization of the incident beam were in the ab plane and the linear vertical (LV) polarization along the c axis of the $\text{BaCo}_2(\text{AsO}_4)_2$ single crystal. The crystal was not oriented in plane. The experimental geometry is shown in Fig. 1(c). The sample was cleaved *in situ* in an ultra-high-vacuum chamber before being transferred into the measurement chamber.

The beam polarization was fixed to either LH or LV during each scan, thus probing the linear dichroism (LD) between the in-plane and out-of-plane directions of the crystal, respectively. The LD measurements were conducted in zero magnetic field at temperatures $T = 300$ K, 200 K, 100 K, 50 K, and 5 K. For the XMCD measurements, a magnetic field of 6 T was applied along the Poynting vector of the incident x-ray beam, which was in the ab plane of the sample. The helicity of the x-ray beam was switched between left (σ^-) and right (σ^+) circular polarization for each of the energy scans. The measurements were carried out both +6 T and −6 T applied magnetic fields to cancel any asymmetry in the background. These measurements were conducted at $T = 50$ K and 5 K.

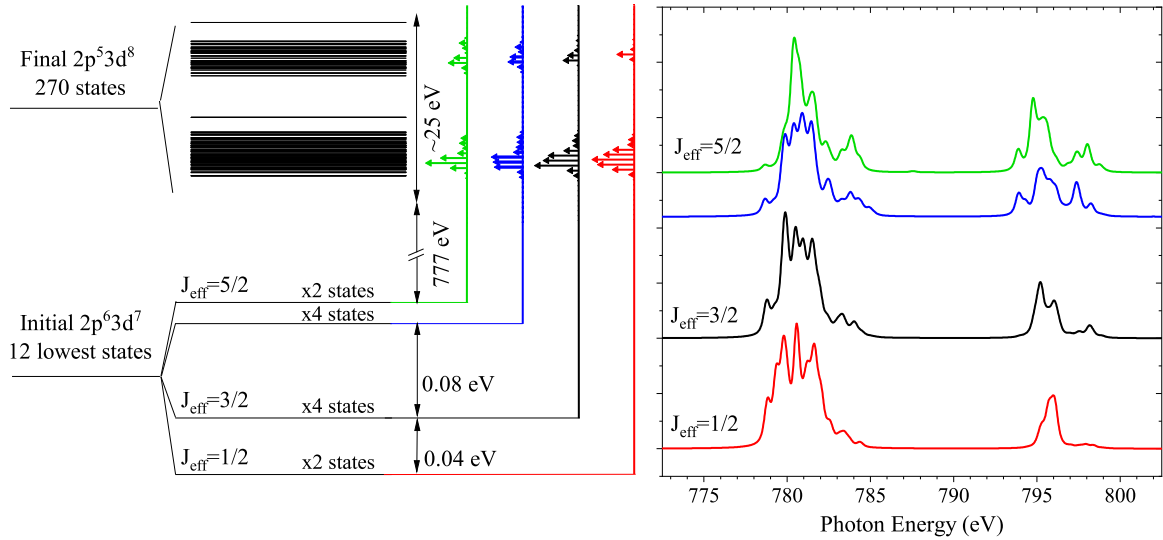


FIG. 2. Left: 12 lowest states depicting the ground state and low lying excited states of a $\text{Co}^{2+} 3d^7$ ion in O_h crystal field. The arrows depict in an x-ray absorption experiment at the $\text{Co } L_{2,3}$ edges the allowed dipole transitions from each initial state into the 270 possible final states split by the $\text{Co } 2p$ spin-orbit coupling. Right: Theoretical spectra corresponding to each starting $J_{\text{eff}} = 1/2, 3/2$, and $5/2$ initial states.

It should be noted that the pink colored transparent crystal of $\text{BaCo}_2(\text{AsO}_4)_2$ with an optical band gap of ≈ 3 eV is highly insulating. Sample charging thus prevents the widely used total electron yield (TEY) method. It is also well known that fluorescence yield (FY) can not be used for a quantitative analysis of the XAS spectra due to distortions of the spectra as a consequence of self absorption effects. Therefore, we used the inverse partial fluorescence yield method (iPFY) developed by the Hawthorn and co-workers [32,33]. They have shown that iPFY method is bulk sensitive and free of saturation effects. To measure iPFY, we used an energy sensitive silicon drift detector (SDD) to record the x-ray emission signal for a wide range of photon energies while scanning the incident x-ray photon energy through the $\text{Co } L_{2,3}$ edges.

For the simulation of the XAS and XMCD spectra, we used the Quanty code [34–37]. The crystal field splittings between the $\text{Co } e_g^\sigma, e_g^\pi$, and a_{1g} orbitals was treated as a fitting parameter, adjusted to match the experimental spectra. The hybridization parameters and the crystal field acting on the oxygen ligands were initially derived from a tight-binding model based on the *ab initio* Wannierized LDA band structure, calculated using FPLO [38,39]. To achieve better agreement with the experimental data, the hybridization parameters were subsequently optimized by tuning them to 84.5% of their initial values.

Field-dependent magnetization was measured using a commercial magnetometer (MPMS3, Quantum Design).

III. RESULTS AND DISCUSSION

A. X-ray absorption

A typical fluorescence yield (FY) intensity map of $\text{BaCo}_2(\text{AsO}_4)_2$ measured as a function of incident photon energy and emission energy is presented in Fig. 3(a). It can be seen that the intensities change significantly when the incident energy crosses the $\text{Co } L_{2,3}$ (778 eV, 792 eV) and $\text{Ba } M_{4,5}$

(784 eV, 798 eV) white lines. As far as emission energies are concerned, one can distinguish the $\text{O } K_\alpha$ (≈ 520 eV), $\text{Ba } M_\alpha$ (≈ 605 eV), $\text{Co } L_\ell$ (≈ 680 eV), and $\text{Co } L_{\alpha,\beta}$ (≈ 780 eV) transitions. Figures 3(b)–3(e) depict the emission spectra for selected incident photon energies corresponding to the pre-Co and Ba edges (760 eV), at the $\text{Co } L_3$ white line (778 eV), at the $\text{Ba } M_5$ white line (784 eV), and after the Co and Ba white lines (820 eV).

Essential for the application of the iPFY method [32,33] is the presence of an emission line with an energy lower than that of the absorption edge of interest. In our case we have access to the $\text{O } K_\alpha$ emission for studying the $\text{Co } L_{2,3}$ absorption spectrum of the $\text{BaCo}_2(\text{AsO}_4)_2$ material. The intensity of this $\text{O } K_\alpha$ emission decreases when the incident photon energy is tuned across the $\text{Co } L_{2,3}$ (and $\text{Ba } M_{4,5}$) absorption edges; see Fig. 4(a). Taking an energy window (or region of interest, ROI) between 480 eV and 560 eV around the $\text{O } K_\alpha$ line as indicated by the gray box in Fig. 3(a), we obtain the gray curve in Fig. 4(a) as the $\text{O } K_\alpha$ PFY. The $\text{Ba } M_\alpha$ emission energy is, however, quite close to that of the $\text{O } K_\alpha$ causing a partial overlap of the emission spectra due to the limited resolution of the SDD instrument. We therefore also have extracted the $\text{O } K_\alpha$ intensity using a Voigt fit as illustrated in Figs. 3(b)–3(e). The resulting $\text{O } K_\alpha$ PFY is plotted as the red curve in Fig. 4(a). The differences between the gray (ROI) and red (Voigt fit) curves are not negligible, which is of relevance for the extraction of the $\text{Co } L_{2,3}$ (and $\text{Ba } M_{4,5}$) XAS spectra.

Figure 4(b) shows the $\text{Co } L_{2,3}$ and $\text{Ba } M_{4,5}$ XAS spectra of $\text{BaCo}_2(\text{AsO}_4)_2$ extracted using various acquisition methods. The red curve is the iPFY [32,33] spectrum based on the $\text{O } K_\alpha$ PFY signal from the Voigt fit to the $\text{O } K_\alpha$ emission line. The gray curve is the iPFY spectrum based on the ROI around the $\text{O } K_\alpha$ emission line. We can clearly observe differences in the extracted XAS spectra. In particular the strongest peaks like the $\text{Ba } M_{4,5}$ suffer from considerable loss of intensity if the $\text{Ba } M_\alpha$ emission enters the iPFY equation. The $\text{Co } L_{2,3}$

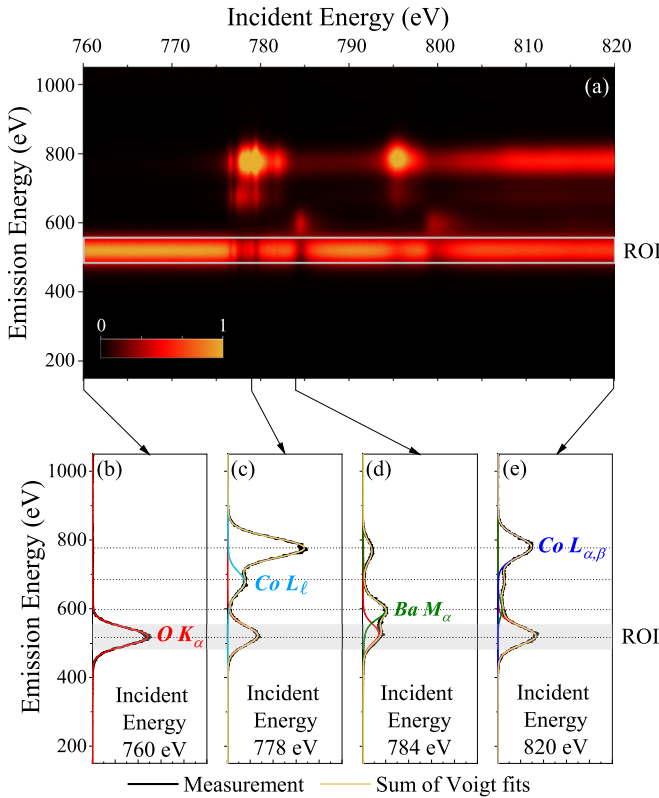


FIG. 3. (a) Fluorescence yield (FY) intensity map of $\text{BaCo}_2(\text{AsO}_4)_2$ as a function of incident photon energy and emission energy, measured at 5 K using LH polarization, and normalized to the incident photon flux. The gray rectangle represents the region of interest (ROI) around the $\text{O } K_\alpha$ emission line used for obtaining the partial fluorescence yield (PFY) spectra shown in Fig. 4. (b)–(e) Fluorescence emission spectra for selected incident photon energies corresponding to the pre-Co and Ba edges (760 eV), at the $\text{Co } L_3$ white line (778 eV), at the $\text{Ba } M_5$ white line (784 eV), and in the post-edge region (820 eV). The red, green, light blue, and dark blue lines are Voigt fits for the $\text{O } K_\alpha$, $\text{Ba } M_\alpha$, $\text{Co } L_\ell$, and $\text{Co } L_{\alpha,\beta}$ emission lines, respectively. The yellow line is the sum of the Voigt fits and black the experimental spectrum.

seems to be less affected, but for the remainder of our study we will utilize the $\text{O } K_\alpha$ PFY signal from the Voigt fit to ensure maximum reliability. For completeness, we show also in Fig. 4(b) the XAS spectra obtained using the $\text{Co } L_{\alpha,\beta}$ PFY (dark blue line) and total FY (black line). Here we can clearly observe strong deviations from the $\text{O } K_\alpha$ iPFY (red line), which can be attributed, among others, to the well-known self-absorption effects in PFY and total FY for ions present in high concentrations in the material. In Fig. 5 we report the temperature dependence of the isotropic $\text{Co } L_{2,3}$ and $\text{Ba } M_{4,5}$ spectra. These spectra were constructed from $(2 * I_{\text{LH}} + I_{\text{LV}})/3$ where I_{LH} and I_{LV} are the iPFY spectra taken with LH and LV polarization, respectively. The observed temperature dependence arises from the thermal population of low-lying excited states as the temperature increases. This is consistent with the expectation that the lowest-lying states for a high-spin Co^{2+} ion in octahedral coordination are split in energy by the $\text{Co } 3d$ SOC constant, which is of the order of 66 meV, i.e., small enough so that within a temperature range of a few hundred Kelvin,

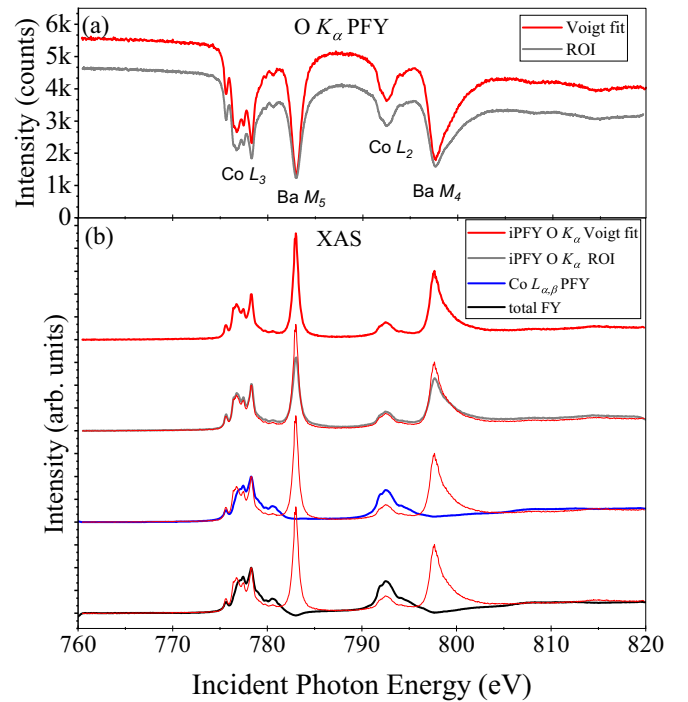


FIG. 4. (a) Comparison between the $\text{O } K_\alpha$ PFY across the $\text{Co } L_{2,3}$ and $\text{Ba } M_{4,5}$ absorption edges using the gray rectangle ROI in Fig. 3(a) and that using a Voigt fit as exemplified in Fig. 3(b). (b) Comparison between the XAS spectra across the $\text{Co } L_{2,3}$ and $\text{Ba } M_{4,5}$ absorption edges obtained using different acquisition methods: $\text{O } K_\alpha$ iPFY using the Voigt fit (red line), $\text{O } K_\alpha$ iPFY using the ROI (gray line), $\text{Co } L_{\alpha,\beta}$ PFY (dark blue line), and total FY (black line).

a nontrivial behavior of the magnetic susceptibility [30] can be measured as well as changes in the spectra as suggested already in Fig. 2. In fact, using parameters as will be described below, we are able to reproduce excellently the experimental data using the full-multiplet configuration-interaction calculations for a CoO_6 cluster. This implies that we have been successful in determining the character of the low-lying states and their energy separations.

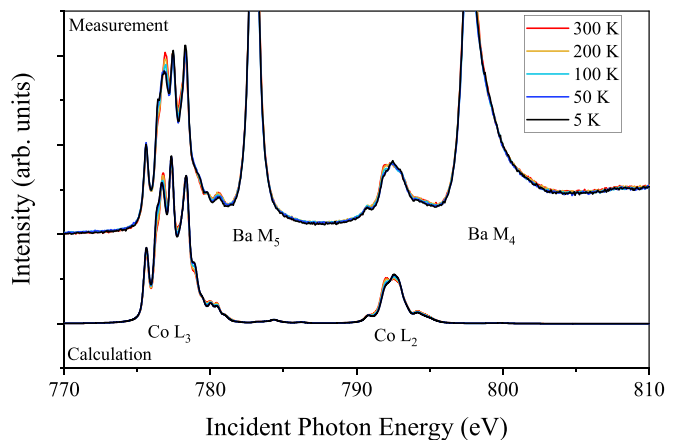


FIG. 5. Comparison between experimental and calculated temperature dependence for the isotropic spectra for $\text{BaCo}_2(\text{AsO}_4)_2$.

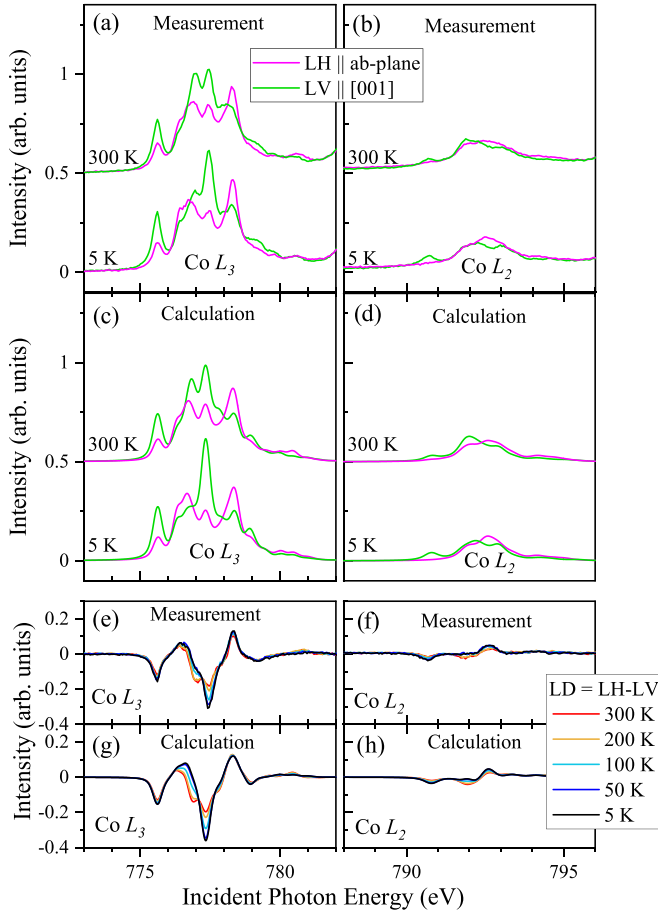


FIG. 6. Top panels: polarization-dependent Co $L_{2,3}$ XAS spectra taken at 5 K and 300 K: (a), (b) experiment and (c), (d) calculation. The incident beam polarization is in the ab plane (LH) and along the [001] direction (LV). Bottom panels: Temperature dependence of the linear dichroic spectra $I_{LD} = I_{LH} - I_{LV}$: (e), (f) experiment and (g), (h) calculation.

B. Linear dichroism

In order to investigate whether crystal fields with lower than octahedral symmetry are present in $\text{BaCo}_2(\text{AsO}_4)_2$, we performed polarization-dependent Co $L_{2,3}$ XAS measurements. Figures 6(a) and 6(b) report the experimental spectra taken at 5 K and 300 K with LH (in the ab plane) and LV (along the [001] direction) polarization. We can observe a significant polarization dependence, which can be taken as an indication that there is a strong distortion of the CoO_6 octahedra in this material. We can also follow this polarization dependence in more detail by plotting the linear dichroism (LD) spectra defined as the difference between the LH and LV spectra. Figures 6(e) and 6(b) reveal that the LD spectra become somewhat smaller in amplitude with increasing temperature but do not vanish at the highest measured temperatures.

To estimate the magnitude of the distortion of the CoO_6 octahedra we set out to simulate the spectra by introducing a trigonal distortion in our configuration-interaction cluster calculations. We limit ourselves to the D_{3d} symmetry as an approximation for the Co^{2+} site symmetry C_3 . The z axis is

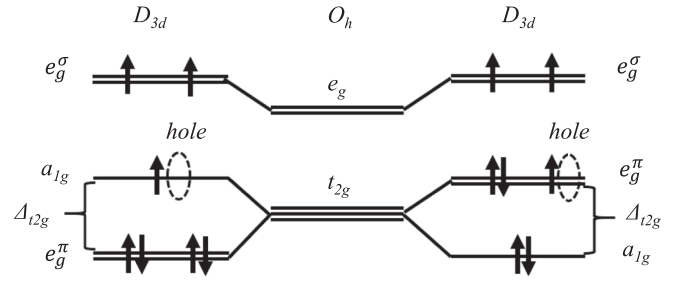


FIG. 7. Schematic diagram showing the energy levels of a d shell in O_h symmetry (center) and their splitting under the influence of a trigonal distortion of D_{3d} symmetry in the absence of SOC. The energy hierarchy of the orbitals under elongation along the crystallographic [001] axis is presented on the left side and that under compression is shown on the right side.

here defined along the [001] crystallographic axis or c axis of the crystal. The energy levels of the d orbitals split into a set of a_{1g} , e_g^π , and e_g^σ orbitals as shown in Fig. 7. Depending on whether the Co^{2+} octahedra undergo an elongation or compression, the t_{2g} hole can reside in the a_{1g} or in the e_g^π orbitals, respectively. Making use of the orbital-polarization sum rule in XAS [41], we can infer that the presence of an a_{1g} hole will lead to a stronger integrated XAS signal when the polarization is along the z axis, i.e., LV polarization. Here we note that the two e_g^σ holes do not lead to a polarization dependence in the integrated XAS intensity. For an e_g^π hole the signal will be stronger with the polarization in the ab plane, i.e., LH polarization. The experiment clearly indicates that the t_{2g} hole is predominately in the a_{1g} orbital, i.e., CoO_6 octahedra are elongated along the z axis.

To be quantitative, we calculate the XAS and the LD spectra with the Quanty code [34–36]. The parameters used can be found in Ref. [42], and the results are plotted in Figs. 6(c) and 6(d) and in Figs. 6(g) and 6(h). We are able to obtain a very good agreement between calculations and measurements for all temperatures, implying that we have indeed found the crystal field and hopping parameters describing the local electronic structure of the Co^{2+} ions in $\text{BaCo}_2(\text{AsO}_4)_2$.

In our calculations, we define the energy of the ionic trigonal splitting as $D_{\text{trig}}^{\text{ion}} = E(e_g^\pi) - E(a_{1g})$; i.e., in this notation, negative values imply elongation and positive values compression of the octahedra. To arrive at the best simulation, we used a value of $D_{\text{trig}}^{\text{ion}} = -116.8$ meV, i.e., the octahedra are elongated and the t_{2g} hole is predominately in the a_{1g} orbital.

In terms of effective crystal fields, i.e., including the effect of hybridization with the O $2p$ ligands, we calculated the total energy levels by setting $\text{SOC} = 0$. For the effective $10Dq^{\text{eff}}$, we set $D_{\text{trig}}^{\text{ion}} = 0$ and find that the difference in energy between the ground state $^4T_1$ with $t_{2g}^5 e_g^2$ configuration and the excited state $^4T_2$ with $t_{2g}^4 e_g^3$ configuration is 0.66 eV; this is the value of $10Dq^{\text{eff}}$. For the effective trigonal distortion $D_{\text{trig}}^{\text{eff}}$, we turn on $D_{\text{trig}}^{\text{ion}} = -116.8$ meV and calculate the energy of the $^4T_1$ term relative to ^4A and ^4E . The energy difference is -114.08 meV, which is thus the magnitude of $D_{\text{trig}}^{\text{eff}}$.

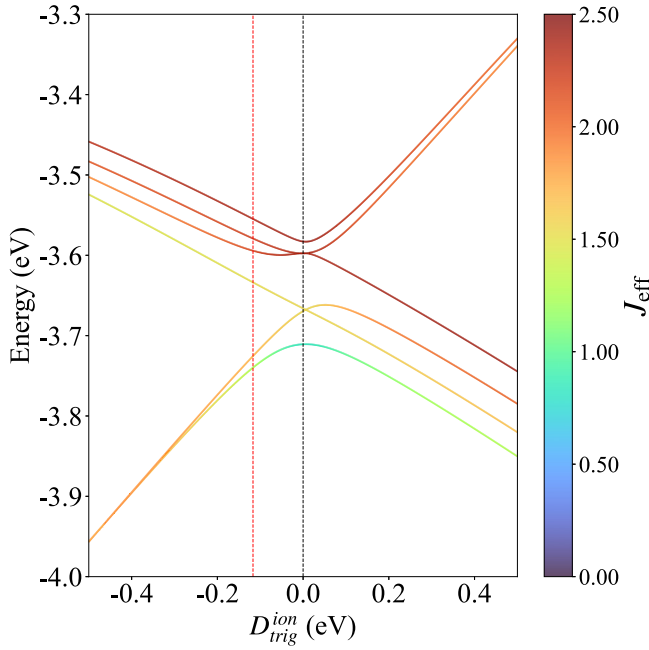


FIG. 8. Energy level diagram showing the evolution of the lowest 12 eigenstates under the influence of SOC and D_{trig}^{ion} . The vertical dashed black line corresponds to $D_{trig}^{ion} = 0$ and vertical dashed red line corresponds to $D_{trig}^{ion} = -116.87$ meV. The color scale indicates the J_{eff} value.

Since the strength of the trigonal distortion we extracted from our experiments is nearly twice the value of the spin-orbit coupling constant of Co^{2+} ions [42], it is important to visualize its impact on the J_{eff} ground state in our material. To this end we use $\mathbf{J}_{eff} = \mathbf{L}_{eff} + \mathbf{S}$, where the $\mathbf{L}_{eff}$ operator is obtained by rotating the orbital basis of the $\mathbf{L}$ operator to the cubic harmonics [43]. The rotation matrix was modified to keep only the t_{2g} subset of the $3d$ eigenorbitals. After projecting out the e_g orbitals, the angular momentum operator is rotated back to the spherical harmonics. As the covalence mixes the Co $3d$ and ligand orbitals, we calculate the expectation value of the $\mathbf{J}_{eff}^2$ operator acting on both the Co $3d$ and ligand d shell, i.e., acting on the total CoO_6 cluster.

The ideal $J_{eff} = 1/2$ is valid when only the t_{2g} orbitals span the Hilbert space, e.g., when the e_g orbitals are completely projected out by making $10Dq$ infinitely large. A value of $J_{eff} = 0.5015$ is obtained for $10Dq = 10$ eV, indicating that this is already close to the ideal situation. However, for the $10Dq^{eff} = 0.66$ eV value used to reproduce the experimental spectra of $\text{BaCo}_2(\text{AsO}_4)_2$, we find $J_{eff} = 0.866$. This is for the undistorted octahedron ($D_{trig}^{ion} = 0$), indicating the strong participation of the e_g orbitals in addition to the t_{2g} . Including the trigonal distortion of $D_{trig}^{eff} = -114.08$ meV found for this material, we arrive at $J_{eff} = 1.302$, a value substantially far from the ideal case. Nevertheless, this state is still doubly degenerate. In Fig. 8 we show the evolution of the energies of the lowest 12 eigenstates as a function of D_{trig}^{ion} together with the colors indicating the J_{eff} values.

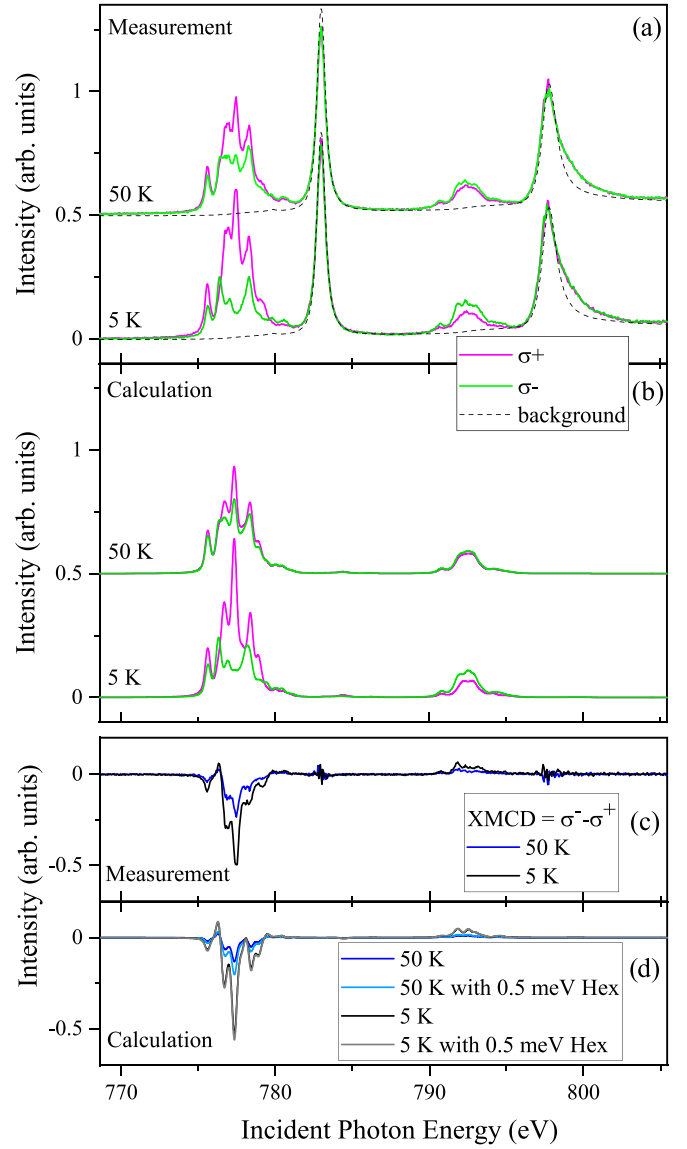


FIG. 9. (a) Experimental and (b) calculated Co $L_{2,3}$ XAS of $\text{BaCo}_2(\text{AsO}_4)_2$ at 5 K and 50 K for positive (σ^+) and negative (σ^-) helicity of the circularly polarized incident beam for an applied field of $B = 6$ T along the ab plane of the sample. (c) Experimental and (d) calculated XMCD signal obtained from the XAS spectra shown in (a) and (b), respectively.

C. X-ray magnetic circular dichroism

To experimentally determine the spin moment $2\langle S_x \rangle$ and orbital moment $\langle L_x \rangle$ of $\text{BaCo}_2(\text{AsO}_4)_2$, we performed XAS measurements at 5 K and 50 K by applying an in-plane magnetic field of 6 T. $\text{BaCo}_2(\text{AsO}_4)_2$ displays a complex magnetic phase diagram for magnetic fields below 1 T [16,19], whereas, under a magnetic field of 6 T, the magnetic moments in the sample are fully aligned in the direction of the applied magnetic field, i.e., in the ab plane (see Fig. 10 in the Appendix). In Fig. 9(a) the XAS measurements performed using σ^+ and σ^- polarization are presented. The XMCD spectra were obtained by taking the difference ($\sigma^- - \sigma^+$), which is shown in Fig. 9(c).

The expectation values $2\langle S_x \rangle$ and $\langle L_x \rangle$ can be obtained by application of XMCD sum rules [44,45] as given in Eq. (1) and Eq. (2):

$$L = \frac{4 \int_{L_{2,3}} (\sigma^+ - \sigma^-) dE}{3 \int_{L_{2,3}} (\sigma^+ + \sigma^-) dE}, \quad (1)$$

$$2S + 7T = 2 \frac{\int_{L_3} (\sigma^+ - \sigma^-) dE - 2 \int_{L_2} (\sigma^+ - \sigma^-) dE}{\int_{L_{2,3}} (\sigma^+ + \sigma^-) dE} N_h. \quad (2)$$

To determine the relevant sum-rule integrals from the spectra, we removed the step-edge and Ba peak contribution by subtracting the background as shown by the dashed line in Fig. 9(a). The spectra were also normalized to the area of the isotropic spectrum, i.e., $(\sigma^+ + \sigma^-)/2$ of the L_3 edge. The magnetic dipole moment T_x is typically a small number (0.012 for the ground state in our cluster calculation) compared to $2\langle S_x \rangle$, and hence will be neglected. Applying the sum rules to the 5 K spectra and using $N_h = 2.89$ from the cluster calculation yields $2\langle S_x \rangle = -1.99$ and $\langle L_x \rangle = -0.78$.

To obtain more reliable results, we first calculated the XMCD spectra [see Fig. 9(b)] using the same parameters as for the LD analysis [42]. It can be seen that the calculated spectra match quite well with the experimental ones. We then extract the desired quantum numbers from the ground state of the cluster. We find $2\langle S_x \rangle = -2.014$ and $\langle L_x \rangle = -0.710$. The difference is less than 10%, indicating that the sum rules work rather well for this compound. This close agreement, indicates that the subtraction of the Ba contribution from the XAS spectra was effective. The total magnetic moment projected on the applied magnetic field direction, i.e., $M_x = |\langle L_x \rangle + 2\langle S_x \rangle|$, is $2.72 \mu_B/\text{Co}$ and $2.82 \mu_B/\text{CoO}_6$ cluster. These values are in reasonable agreement with the saturation magnetization of $2.92 \mu_B/\text{Co}$ from the $M(H)$ in-plane measurements presented in Fig. 10. The effective g factors obtained from the relation $M = gM_{J_{\text{eff}}}$ with $M_{J_{\text{eff}}} = 1/2$ from our cluster calculations using an applied field of 6 T are $g_{ab} = 5.64$ and $g_c = 2.49$. These values compare well with $g_{ab} = 5.0$ and $g_c = 2.5$ – 2.7 from previously reported experiments [46,47]. The significant difference between g_{ab} and g_c contributes to the strong magnetocrystalline anisotropy observed in this material. Concerning the 50 K data, we notice that the simulation produces a smaller XMCD effect than the experiment. This can be resolved by adding an exchange field of 0.5 meV to the 6 T magnetic field to represent short-ranged inter-Co exchange interactions.

D. Discussion

Using polarization-dependent XAS, we have experimentally determined the energy of the trigonal splitting $D_{\text{trig}}^{\text{eff}} = -114.08 \text{ meV}$ in $\text{BaCo}_2(\text{AsO}_4)_2$. The value of $|D_{\text{trig}}^{\text{eff}}|$ found here is in good agreement with that reported in recent Raman spectroscopy studies [48]. Note that the negative sign of $D_{\text{trig}}^{\text{eff}}$ in our studies is due only to our notation $D_{\text{trig}}^{\text{ion}} = E(e_g^\pi) - E(a_{1g})$, which is the opposite of what is reported in Ref. [48]. This value is nearly twice the value of the atomic spin-orbit coupling constant of 66 meV of Co^{2+} ions. This implies significant deviations from the ideal $J_{\text{eff}} = 1/2$ ground state desired for the formation of a pure Kitaev quantum spin liquid.

The magnitude of trigonal distortion obtained here is consistent with *ab initio* density-functional theory, which takes into account realistic crystal-field distortions [49]. *Ab initio* calculations of effective spin models for $\text{BaCo}_2(\text{AsO}_4)_2$ yield a negligible Kitaev term [50], whereas experimental estimates of the Kitaev term vary in both sign and magnitude [20–23]. A time-domain terahertz spectroscopy study reported a broad magnetic continuum in another part of the phase diagram, i.e., in a magnetic field of 4 T applied in the out-of-plane direction. The magnetic continuum is found to be suppressed below T_N , and this behavior is considered to be compatible with the spin-liquid models describing fractionalized excitation [51].

On the other hand, application of hydrostatic pressure changes relative stability of the different phases in the field-temperature phase diagram, but it does not suppress magnetic order in any prominent way [19,52]. The elongated trigonal distortion of CoO_6 observed in our experiments suggests that uniaxial compressive strain applied along the c axis or a stretching of ab plane might be a more effective method to suppress the conventional long-range magnetic order in $\text{BaCo}_2(\text{AsO}_4)_2$, although c -axis pressure initially increases T_N [19]. A suppression of T_N through strain engineering of another honeycomb lattice $\text{Cu}_3\text{Co}_2\text{SbO}_6$ in thin films form has been reported [53].

IV. CONCLUSIONS

We have successfully carried out polarization dependent soft x-ray absorption spectroscopy on the Kitaev candidate honeycomb lattice compound $\text{BaCo}_2(\text{AsO}_4)_2$ using the iPFY method, which is the most reliable manner to collect spectra suitable for quantitative analysis on highly insulating samples. Our results indicate a sizable trigonal distortion causing a splitting of $\approx -114.08 \text{ meV}$ in this compound. The observation of strong temperature dependence of LD confirms the thermal population of excited states in the temperature regime above 100 K. From the XMCD sum rules and cluster-model analysis of the spectra, we obtain the total in-plane magnetic moment of 2.72 – $2.77 \mu_B/\text{Co}$, which is in reasonable agreement with the saturation value of in-plane magnetic moment obtained from the magnetization measurement. Our findings suggest that the application of an uniaxial strain is a possible route to possibly tune the system into a quantum spin liquid regime.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [40].

APPENDIX

To compare the total in-plane magnetic moment M_x obtained from the XMCD sum rules with that from the magnetization measurements, we performed $M(H)$ with the magnetic field H applied parallel to the ab plane. The measurements were performed at $T = 2$ K. In zero applied magnetic field, $\text{BaCo}_2(\text{AsO}_4)_2$ orders antiferromagnetically below $T_N \approx 5.4$ K into an incommensurate helical spin structure derived from the double zigzag type of order [46,54]. Upon application of the in-plane magnetic field, the compound goes through a series of complex phases [16,19], and finally reaches the full spin polarized state of the Co^{2+} ions in the applied magnetic field $\mu_0 H \approx 2$ T as can be seen from Fig. 10. Above 2 T, the $M(H)$ increases due to the Van Vleck paramagnetic contribution, which is marked by the red dashed line in Fig. 10. After subtracting the Van Vleck contribution, we obtain a saturation in-plane magnetic moment of $2.92 \mu_B/\text{Co}$.

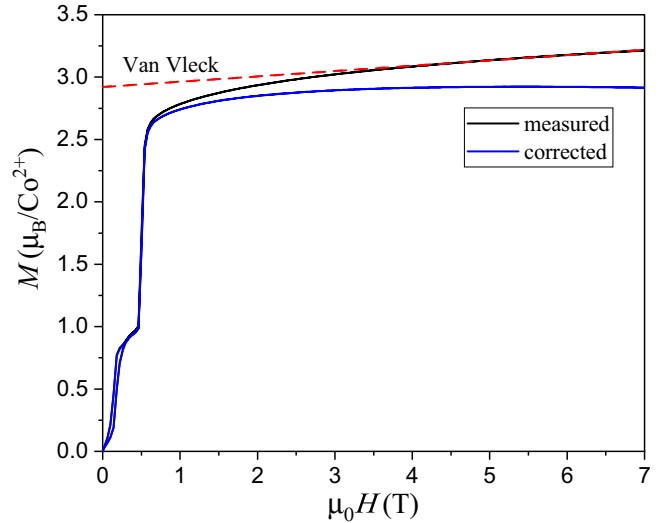


FIG. 10. Magnetization $M(H)$ measured at $T = 2$ K with the applied magnetic field parallel to the ab plane. The dashed red line represents the Van Vleck contribution. The legend label “measured” shows as measured $M(H)$ curve, and “corrected” represents the $M(H)$ data after correcting for the Van Vleck contribution.

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