

Quantum spin ladder with ferromagnetic rungs in $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$

Rodolfo A. Rangel Hernandez ¹, Kirill Yu. Povarov ², Sergei Zvyagin ², Oleg I. Siidra ³,
Alexander A. Tsirlin ^{1,*} and Victoria A. Ginga ^{1,†}

¹*Felix Bloch Institute for Solid-State Physics, University of Leipzig, 04103 Leipzig, Germany*

²*Dresden High Magnetic Field Laboratory (HLD-EMFL) and Würzburg-Dresden Cluster of Excellence ctd.qmat, Helmholtz-Zentrum Dresden-Rossendorf (HZDR), 01328 Dresden, Germany*

³*Department of Crystallography, St. Petersburg State University, 199034 St. Petersburg, Russia*



(Received 26 March 2026; revised 2 June 2026; accepted 10 June 2026; published 6 July 2026)

We introduce $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ as a rare example of a spin-ladder magnet with ferromagnetic interactions on the rungs. Its magnetic response is studied through measurements of temperature-dependent magnetic susceptibility and heat capacity, field-dependent magnetization, and electron spin resonance spectroscopy. These experiments are complemented by density-functional-theory calculations combined with the construction of maximally localized Wannier functions and an analysis of the relevant superexchange pathways. Quantum Monte Carlo simulations are employed to model thermodynamic properties and to quantitatively determine the magnetic exchange parameters. Our combined approach identifies $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ as a two-leg spin-ladder system with ferromagnetic rungs ($J' \approx -208$ K) and antiferromagnetic legs ($J \approx 258$ K). These interactions of similar magnitude arise from remarkably different superexchange pathways, with the Cu–Cu distance along the leg being almost twice as long as the respective distance along the rung. The antiferromagnetic leg coupling represents the strongest oxygen-mediated long-range superexchange in a Cu^{2+} compound and sets a benchmark for the role of complex superexchange pathways in quantum magnets.

DOI: [10.1103/nwlh-vnyp](https://doi.org/10.1103/nwlh-vnyp)

I. INTRODUCTION

Low-dimensional quantum magnets have been the subject of extensive theoretical and experimental interest for several decades. These materials display a variety of unconventional ground- and excited-state properties that emerge from strong quantum fluctuations and competing interactions [1–4]. Transition-metal compounds containing Cu^{2+} ions with a $3d^9$ configuration provide numerous realizations of such physics, since subtle changes in the local coordination environment can drastically modify the effective magnetic dimensionality and stabilize diverse magnetic ground states [3,5–7]. An effective strategy to tune the magnetic dimensionality in Cu-based systems involves the incorporation of stereochemically active lone-pair cations such as Bi^{3+} or Te^{4+} [8,9]. Their $6s^2$ (or $5s^2$) lone pairs introduce asymmetric local distortions that modify Cu–O–Cu bond geometries and thereby reshape the underlying superexchange network.

Several of the copper-based $S = 1/2$ materials adopt the spin-ladder geometry. The Cu^{2+} ions form networks of corner- or edge-sharing CuO_4 plaquettes that further assemble into two-leg or multileg ladders, resulting in markedly different magnetic ground states. Even-leg ladders feature a

finite spin gap, whereas odd-leg ladders behave similarly to single Heisenberg chains and feature gapless spin excitations [2,8,10]. One common scenario of the ladder formation is the condensation of CuO_4 plaquettes via single oxygen atoms, as in SrCu_2O_3 [10], CaCu_2O_3 [11], and $\text{La}_2\text{Cu}_2\text{O}_5$ [12] whose magnetic behavior is well described by the model of a two-leg spin ladder with dominant rung–leg interactions mediated by Cu–O–Cu superexchange, and no competing exchange pathways.

In contrast, BiCu_2PO_6 [13], which crystallizes in the orthorhombic space group $Pnma$ and comprises ribbons of edge-sharing CuO_4 plaquettes forming coupled two-leg ladders along the b axis, stands out as one of the few known $S = 1/2$ ladder systems with magnetic frustration caused by sizable antiferromagnetic (AFM) next-nearest-neighbor (NNN) exchanges along the ladder legs [9,14]. These NNN pathways compete with nearest-neighbor interactions, which are also AFM, and give rise to a peculiar energy spectrum that features singlet bound modes [15] and the quasiparticle-continuum level repulsion [16], along with a series of field-induced phases [17], including the quantum soliton lattice [18]. Magnetic anisotropy in the form of Dzyaloshinskii–Moriya interactions is known to be important for the quantitative description of this material [19–21]. On the structural level, one of its peculiarities is the importance of long-range superexchange via two consecutive oxygen atoms that underlies the rung couplings as well as the frustrating NNN couplings along the legs [9].

In the following, we introduce another member of the family of BiCu-based spin-ladder materials, the previously unexplored $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ [22]. The compound $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$,

*Contact author: altsirlin@gmail.com

†Contact author: victoria.ginga@uni-leipzig.de

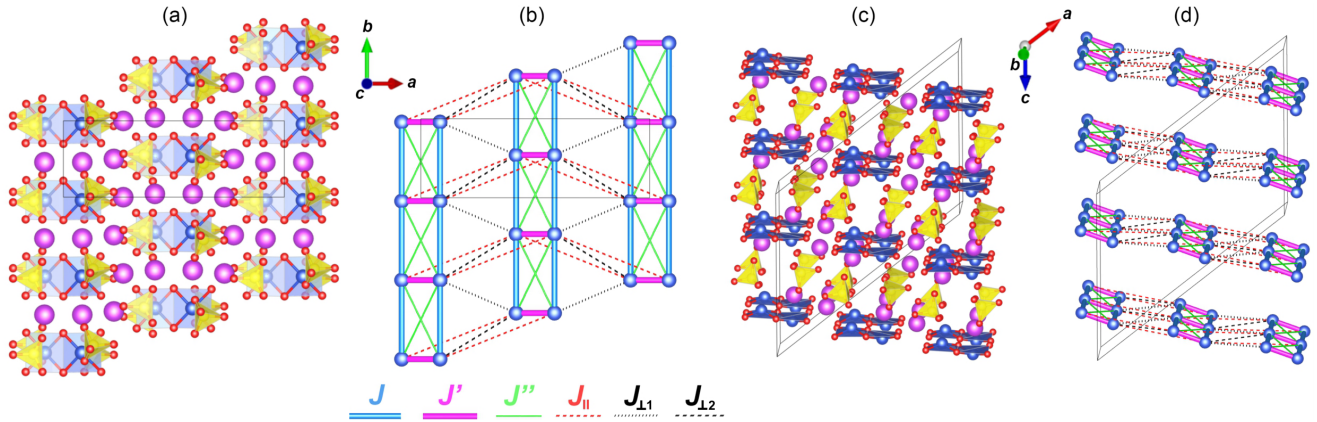


FIG. 1. General projections of the crystal structure and spin lattice of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ along c (a), (b) and within the ac plane (c), (d) (Cu: blue balls, Bi: purple balls, CuO_4 plaquettes are shown in blue, $(\text{SO}_4)^{2-}$ tetrahedra are yellow). The spin lattice comprises the leg J (cyan), rung J' (pink), diagonal J'' (green), as well as the interladder $J_{||}$ (red), $J_{\perp 1}$ (black dotted) and $J_{\perp 2}$ (black dashed) couplings.

although sharing the same nominal stoichiometry, adopts a distinct crystal structure featuring corner-sharing CoO_5 pyramids. The latter form antiferromagnetic chains with a weak ferromagnetic component in the ordered state below approximately 20 K attributed to spin canting [23]. The $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ compound turns out to show a very different magnetic behavior. Our experimental characterization of the magnetic response of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ combines measurements of the temperature-dependent magnetic susceptibility and heat capacity, field-dependent magnetization, and electron spin resonance spectroscopy. Complementing these measurements, we carry out density functional theory (DFT) band-structure calculations and construct Wannier functions (WFs). This allows us to determine the relevant exchange pathways through a mapping procedure. Quantum Monte Carlo simulations are then employed to model thermodynamic properties and quantitatively extract the underlying magnetic couplings. We show that $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ is well described by a model of the two-leg spin ladder with AFM legs and ferromagnetic (FM) rungs (Fig. 1). Long-range superexchange plays a crucial role in the formation of spin ladders, with the remarkably strong exchange couplings in excess of 200 K despite large separations between the Cu^{2+} ions.

II. METHODS

Previously, $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ was prepared by a high-pressure high-temperature reaction of CuSO_4 and BiOCl [22]. In the present work, polycrystalline samples of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ were prepared from CuSO_4 and BiOCl precursors mixed in a molar ratio of 1:2.2. The thoroughly ground precursor mixtures were sealed under vacuum in quartz ampoules and heated following the same temperature profile (including heating and cooling rates) as described in Ref. [22], but without applying external pressure. Two independent synthesis batches of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ were prepared and investigated. Their crystal structure and phase composition were verified by collecting high-resolution x-ray diffraction (XRD) data at 80 K and 298 K (see Supplemental Material [24]) at the ID22 beamline of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France using the wavelength of

0.35433 Å and the multianalyzer detector setup [25]. The crystal structure was refined by the Rietveld method using JANA2006 software [26].

Rietveld refinements against the high-resolution XRD data in Fig. 2 consistently reveal the presence of 8–10 wt. % of the nonmagnetic impurity phase $\text{Bi}_{28}\text{O}_{32}(\text{SO}_4)_{10}$ [27]. All measurements shown below were performed on the sample that did not contain any further impurity phases. The sample from the second batch additionally showed several very weak reflections that could be tentatively assigned to the unreacted BiOCl and to $\text{Cu}(\text{OH})\text{Cl}$, which might form as a secondary phase during the synthesis. While the latter impurity is magnetic, we did not observe any additional anomalies in the

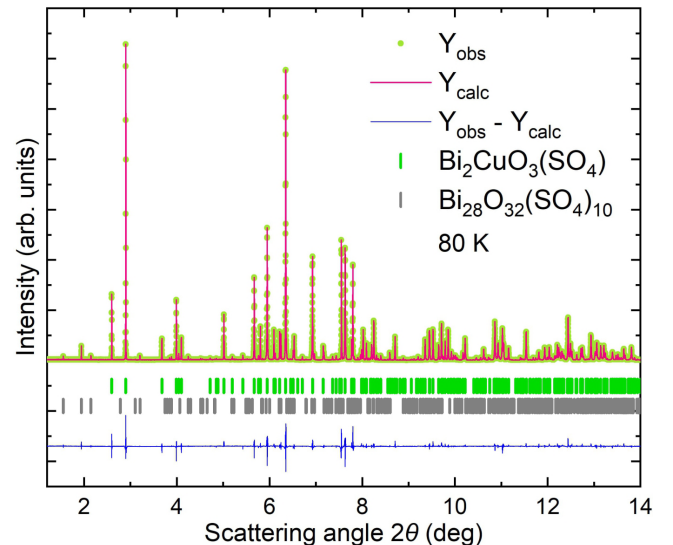


FIG. 2. Crystal structure refinement of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ in the $C2/c$ space group by the Rietveld method from the synchrotron powder data at 80 K (ID22, ESRF). Experimental data are shown as green circles (Y_{obs}), the calculated profile is shown by the red solid line (Y_{calc}), and the difference curve ($Y_{\text{obs}} - Y_{\text{calc}}$) is displayed as a blue line at the bottom. Vertical tick marks indicate the Bragg reflection positions of the main phase $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ (green) and the impurity phase $\text{Bi}_{28}\text{O}_{32}(\text{SO}_4)_{10}$ (gray).

magnetization data. All features reported in this work are well reproducible across the two batches [24] and should be intrinsic to $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$.

Magnetic susceptibility $\chi(T)$ was measured on an 80.5 mg polycrystalline sample of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$. The data were collected in applied magnetic fields of 0.1 T, 1 T, 2.5 T, and 5 T over the temperature range of 2–300 K using a Quantum Design MPMS-XL magnetometer. For temperature-dependent magnetization measurements, both zero-field-cooled (ZFC) and field-cooled (FC) protocols were implemented. Field-dependent magnetization was additionally recorded in magnetic fields up to 7 T.

Heat-capacity $C_p(T)$ measurements were carried out on a pressed pellet (10.1 mg) between 1.9 K and 200 K in external magnetic fields of 0 T, 1 T, 2.5 T, 5 T, and 9 T using the heat-capacity option of the Physical Property Measurement System (PPMS, Quantum Design).

High-frequency electron spin resonance (ESR) measurements were performed at the Dresden High Magnetic Field Laboratory (Hochfeld Magnetlabor-Dresden), using a tunable-frequency spectrometer (similar to the one described in Ref. [28]) equipped with a 16 T superconducting magnet. We employed VDI microwave-chain sources (product of Virginia Diodes, Inc., USA) as the radiation source (100 – 500 GHz frequency range) and a hot-electron n-InSb bolometer (product of QMC Instruments Ltd., UK), operated at 4.2 K, as a detector. We used 2,2-diphenyl-1-picrylhydrazyl (DPPH) as a standard frequency-field marker.

To quantify individual magnetic exchange interactions, scalar-relativistic DFT band-structure calculations were carried out using the FPLO code [29] and the Perdew-Burke-Ernzerhof (PBE) exchange–correlation potential [30]. With PBE calculations, we identified relevant magnetic orbitals and evaluated hopping parameters t_i via a fit with an effective one-orbital tight-binding model constructed from WFs centered on the Cu sites [31]. The use of the WF approach allows for a well-defined evaluation of the hopping integrals t_i and yields a transparent representation of the underlying magnetic orbitals. The resulting hopping parameters t_i provide a microscopic measure of the orbital overlap along individual superexchange pathways. The respective contribution to the exchange couplings gauges kinetic superexchange and elucidates the dominant mechanism of each magnetic coupling in the material. The hopping parameters are introduced into a Hubbard model with the effective on-site Coulomb repulsion potential $U_{\text{eff}} = 8.5$ eV. For half-filled bands in the strongly correlated regime ($t_i \ll U_{\text{eff}}$), the Hubbard model can be further reduced to a Heisenberg model, yielding the antiferromagnetic contributions to the exchange couplings as $J_i^{\text{AFM}} = 4t_i^2/U_{\text{eff}}$.

The ferromagnetic contribution to the superexchange interaction arises from mechanisms not captured within the one-orbital model used in the aforementioned analysis. In cuprate systems, such ferromagnetic exchange is commonly attributed to Hund’s coupling on the ligand sites [32]. As an alternative approach, the magnetic exchange interactions were evaluated by explicitly accounting for strong electronic correlations within the mean-field DFT + U method. Correlations in the Cu $3d$ shell were treated with standard parameters $U_{\text{Cu}} = 8.5$ eV and $J_{\text{Cu}} = 1$ eV and double-counting correction in the atomic limit [33,34]. The individual

TABLE I. Refined crystallographic parameters for $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ in the $C2/c$ space group at 80 K and 298 K.

Parameter	80 K	298 K
a (Å)	19.962(1)	20.037(1)
b (Å)	5.381(1)	5.394(1)
c (Å)	14.098(1)	14.148(1)
β (°)	128.389(1)	128.397(1)
V (Å ³)	1186.881(6)	1198.467(6)
R_1 (%)	1.55	1.79

exchange constants J_i of the Heisenberg Hamiltonian,

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \mathbf{S}_j, \quad (1)$$

where $S = 1/2$ and the summation is over bonds, were obtained by a mapping procedure using total energies of four collinear spin configurations [35]. In all calculations, experimentally determined lattice parameters and atomic positions were employed. Total energies were evaluated using a k -point mesh comprising up to 64 points in the first Brillouin zone.

Thermodynamic properties were further evaluated using quantum Monte Carlo (QMC) simulations performed with the loop algorithm [36] of the ALPS simulation package [37]. Simulations of the magnetic susceptibility were done for finite lattices with periodic boundary conditions. Isolated spin ladders with the length of $L = 32$ sites were used. This length is sufficient to eliminate finite-size effects for thermodynamic properties within the temperature range under investigation. Additionally, we performed simulations for a two-dimensional spin lattice with zigzag couplings between the spin ladders and used scaling properties of the spin stiffness to determine the critical value of the interladder coupling that causes magnetic ordering [38].

III. RESULTS

A. Crystal structure

High-resolution XRD data confirm the structural model of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ with the monoclinic space group $C2/c$ [22]. No peak splittings or superstructure reflections could be observed at either 80 K or 298 K. The refined crystallographic parameters for $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ in the $C2/c$ space group at 80 K and 298 K are summarized in Table I.

All lattice parameters increase slightly upon warming, indicating a moderate thermal expansion of the structure. A tentative estimate of the linear thermal-expansion coefficients returns $\alpha_a \approx 18 \times 10^{-6} \text{ K}^{-1}$, $\alpha_b \approx 11 \times 10^{-6} \text{ K}^{-1}$, and $\alpha_c \approx 17 \times 10^{-6} \text{ K}^{-1}$, indicating predominant thermal expansion within the ac plane. This anisotropic thermal expansion highlights quasi-one-dimensional (1D) nature of the material where spin-ladder units occur along the b direction of the crystal structure (Fig. 1).

The Cu site is surrounded by four oxygen atoms forming a distorted CuO_4 square with the Cu–O distances of 1.91 – 1.99 Å, further capped by two apical O^{2-} ligands with the longer distances of 2.26 Å and 2.76 Å, respectively [24]. Together, they define a distorted $[\text{CuO}_4\text{O}_2]$ octahedron.

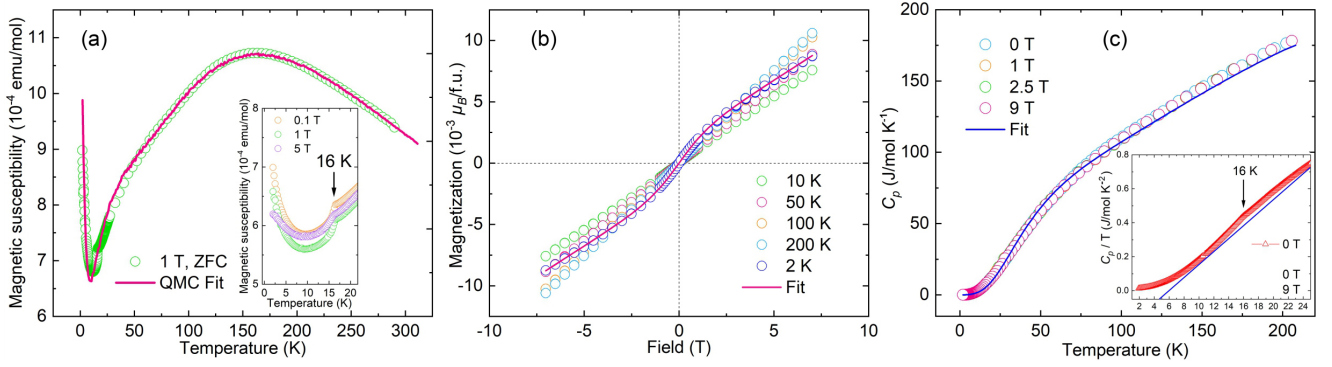


FIG. 3. (a) Temperature-dependent magnetic susceptibility of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ measured in the applied field of 1 T with the QMC fit described in the text. The inset shows the low-temperature region of $\chi(T)$ at 0.1 T, 1 T, 5 T and highlights the anomaly at 16 K. (b) Field-dependent magnetization of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ measured at 2 K, 10 K, 50 K, 100 K, and 200 K with the mean-field fit described in the text. (c) Temperature-dependent specific heat $C_p(T)$ of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ measured at 0 T, 1 T, 2.5 T, and 9 T. The colored circles show the raw data, the blue line represents the estimation of phonon contribution using the model with two Debye temperatures, as explained in the text.

The equatorial planes CuO_4 of these $[\text{CuO}_4\text{O}_2]$ octahedra are linked via edge sharing, effectively forming Cu_2O_6 dimers with the intradimer Cu–Cu distance of 2.79 Å. These dimers are arranged parallel to each other (Fig. 1) and form spin ladders, as we show by the microscopic analysis in Sec. III F.

The dimers are linked via Bi^{3+} ions that feature four shorter Bi–O bonds of 2.23 – 2.37 Å and several further bonds that all exceed 2.65 Å. Such an asymmetry is typical for Bi^{3+} with its stereoactive $6s^2$ lone pair. One interesting feature of the material is that the shorter Bi–O bonds are always directed toward the CuO_4 plaquettes, whereas the longer bonds point toward the SO_4 tetrahedra. This leaves the tetrahedra rather weakly bonded between the planes formed by the CuO_4 plaquettes and Bi^{3+} ions [24]. Indeed, the refined crystal structure shows much higher atomic displacement parameters for the oxygen atoms of the SO_4 tetrahedra ($U_{\text{iso}} = 0.028 \text{ Å}^2$) compared to the oxygen atoms of the CuO_4 plaquettes ($U_{\text{iso}} = 0.012 \text{ Å}^2$) at 298 K. Both values are reduced on cooling. At 80 K, they become 0.009 and 0.005 Å^2 , respectively, indicating that any disorder of the SO_4 tetrahedra is dynamic in nature.

B. Magnetic susceptibility

Temperature-dependent magnetic susceptibility $\chi(T)$ of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ exhibits a broad maximum, characteristic of low-dimensional quantum spin systems [Fig. 3(a)]. The low-temperature upturn below 20 K is commonly associated with paramagnetic contributions from dilute impurities or defects, as further verified by ESR (Sec. III D). The position of the maximum, $T_{\text{max}} \approx 156 \text{ K}$, shows only a weak field dependence and reflects the dominant exchange energy scale in the system. Magnetic susceptibility curves measured at different fields coincide, except in the region below 10 K where the impurity contribution becomes prominent. The small anomaly observed in the magnetic susceptibility near 16 K is fully reproducible across different synthesis batches [24] and likely reflects the onset of long-range magnetic order. We did not detect any magnetic impurity that could be responsible for this anomaly.

The broad susceptibility maximum is typical for various low-dimensional spin systems. Therefore, we start analyzing

the data using simple models with known analytical expressions for the magnetic susceptibility. The dimers of Cu atoms in the crystal structure (Fig. 1) suggest a possible description in terms of spin dimers with

$$\chi_{\text{dimer}}(T) = \frac{N_A g^2 \mu_B^2}{k_B T} \cdot \frac{1}{3 + e^{J/k_B T}} \quad (2)$$

Alternatively, the spin-chain model can be used:

$$\chi_{\text{chain}}(T) = \frac{N_A g^2 \mu_B^2}{4k_B T} \frac{1 + \sum_{n=1}^5 N_n/t^n}{1 + \sum_{n=1}^6 D_n/t^n}, \quad (3)$$

where $t = k_B T/J$ and N_n and D_n are coefficients given in Ref. [39]. However, none of these models reproduce the shape of the experimental susceptibility maximum, even if supplied with additional corrections for the temperature-independent and Curie-like impurity contributions (see Fig. S3 [24]). In Sec. III F, we use a DFT-based microscopic magnetic model to fit the susceptibility curve. Before presenting these results, we consider in more detail the nature of the low-temperature upturn that has to be taken into account when fitting the susceptibility data.

C. Magnetization

Magnetization curves reveal no field-induced magnetic transition. For temperatures above 10 K, the magnetization exhibits a strictly linear dependence on the applied magnetic field, consistent with a predominantly paramagnetic or antiferromagnetic response. A deviation from the ideal paramagnetic behavior of $M(H)$ is visible at 2 K [Fig. 3(b)]. This 2 K magnetization curve is the only one that shows discernible Brillouin-type nonlinearity in the low-field range (–2.5 T to 2.5 T). As expected, such impurity-related contributions diminish rapidly with increasing temperature.

To quantify this low-temperature behavior, we fitted $M(H)$ with a combination of the linear term and Brillouin function B_S ,

$$M(B) = f \cdot N_A g \mu_B S \cdot B_S \left(\frac{S g \mu_B B}{k_B T} \right) + \chi_m \cdot B, \quad (4)$$

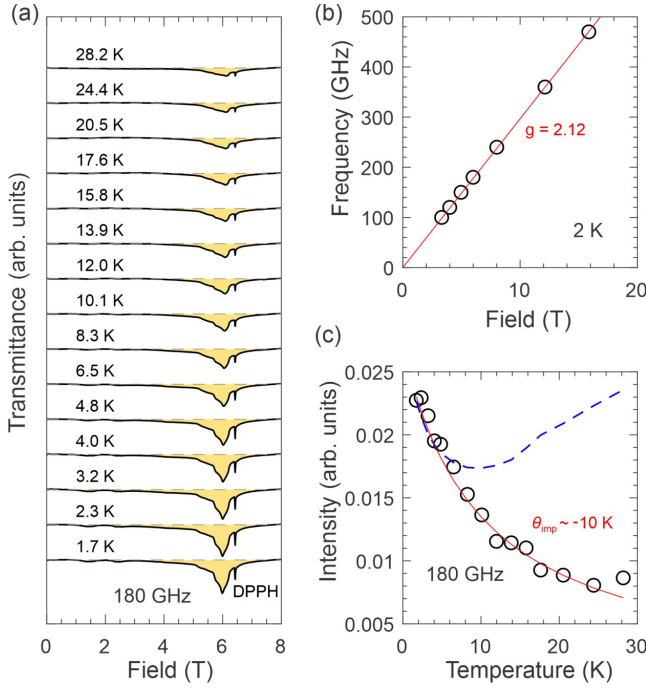


FIG. 4. (a) Exemplary ESR lines at different temperatures at 180 GHz. Shading highlights the area under the curve. The sharp spikes in the spectra reflect the ESR signal of the DPPH marker. (b) Frequency-field diagram at 2 K. Solid line corresponds to $g = 2.12$. (c) ESR line intensity at 180 GHz. Symbols represent the data, solid line is a Curie-Weiss fit. Dashed line is the bulk magnetic susceptibility scaled to match the ESR line intensity at low temperatures.

where f stands for the percentage of paramagnetic impurities present in the sample. Using $S = \frac{1}{2}$, one arrives at an unrealistically high g -value of 5.55 that indicates ferromagnetic correlations between the impurity spins. Such correlations can be taken into account on the mean-field level by introducing an effective field $B + \lambda M$ into the Brillouin function,

$$M(B) = f \cdot N_A g \mu_B S \cdot B_S(a(B + \lambda M)) + \chi_m \cdot B, \quad (5)$$

with $a = Sg\mu_B / k_B T$. This procedure results in a more realistic $g = 2.10$ and $f = 0.11\%$.

The diminutively small amount of impurities inferred from this analysis leads to the susceptibility of about $\chi = 2 \times 10^{-4}$ emu/mol at 2 K, which is several times smaller than the experimental value of 9×10^{-4} emu/mol. We thus conclude that the nonlinearity in $M(H)$ arises from a secondary impurity contribution due to trace amounts of spins with weak ferromagnetic correlations. By contrast, the low-temperature upturn of the magnetic susceptibility is mainly due to antiferromagnetically correlated impurity spins, as we show in the following.

D. Electron spin resonance

The exemplary ESR lines at 180 GHz measured on a pressed pellet of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ are shown in Fig. 4(a). The single observed transmittance dip has a relatively complicated profile, as typical for powder samples, and its amplitude is strongly temperature-dependent. The line at this frequency is centered around the resonant field of 6 T at all

temperatures. The low-temperature frequency-field diagram of the ESR response displayed in Fig. 4(b) suggests that this resonance can be described with an effective powder-averaged g -factor of 2.12(2), which is a typical value for Cu^{2+} ions. For paramagnetic substances, the intensity of the ESR signal (corresponding to the area under the line) is proportional to the magnetization. Thus, it is instructive to compare the temperature-dependent ESR intensity to the bulk magnetic susceptibility. This comparison is shown in Fig. 4(c) for the 180 GHz data. The area estimate is done by numerical integration, with the DPPH contribution excluded. The observed ESR intensity clearly displays a Curie-Weiss behavior (solid line). This is a strong indication of the impurity subsystem being the source of the ESR absorption and the low-temperature upturn in $\chi(T)$.

By describing the ESR intensity with a Curie-Weiss law, $\chi_{\text{imp}} = C_{\text{imp}} / (T - \theta_{\text{imp}})$, we find $\theta_{\text{imp}} \simeq -10$ K and thus directly confirm that the dominant impurity contribution originates from antiferromagnetically coupled impurity spins. This dominant impurity subsystem therefore differs from the weak Brillouin-like nonlinearity observed only in the 2 K $M(H)$ curve, which likely arises from a much smaller concentration of weakly interacting or nearly free spins. A possible microscopic origin of the dominant impurity subsystem is related to structural imperfections producing vacancies within the spin ladders of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$. In gapped dimerlike systems, such vacancies locally break singlet correlations and effectively release uncompensated ($S = \frac{1}{2}$) moments that behave as impurity spins with weak FM or AFM interactions depending on the separation between the defects [40–42].

It is worth noting that only the impurity contribution associated with the low-temperature susceptibility upturn could be observed in our ESR measurements. In a gapped spin-ladder system, the ESR response of the intrinsic Cu subsystem is expected to be weak and thermally activated because it originates from excited triplet states above the singlet ground state. In addition, magnetic anisotropies may modify the resonance conditions, while powder averaging further reduces the observable signal intensity. These effects naturally explain why the ESR response of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ is dominated by impurity-related absorption.

E. Heat capacity

To verify the susceptibility anomaly at 16 K, heat capacity of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ was measured in zero and applied magnetic fields. The resulting data are shown in Fig. 3(c). No field-induced effects are observed within the experimental resolution. Likewise, heating and cooling measurements reveal no substantial differences, indicating the absence of thermal hysteresis in the studied temperature range.

The zero-field data were fitted in the range of 2–200 K using an empirical model with two Debye temperatures,

$$C_p(T) = 9R \sum_{n=1}^2 A_n \left(\frac{T}{\theta_{Dn}} \right)^3 \int_0^{\theta_{Dn}/T} \frac{x^4 e^x}{(e^x - 1)^2} dx, \quad (6)$$

where R is the universal gas constant, and A_1 and A_2 are dimensionless factors describing the relative weight of each Debye contribution to the total specific heat. The fit yields the

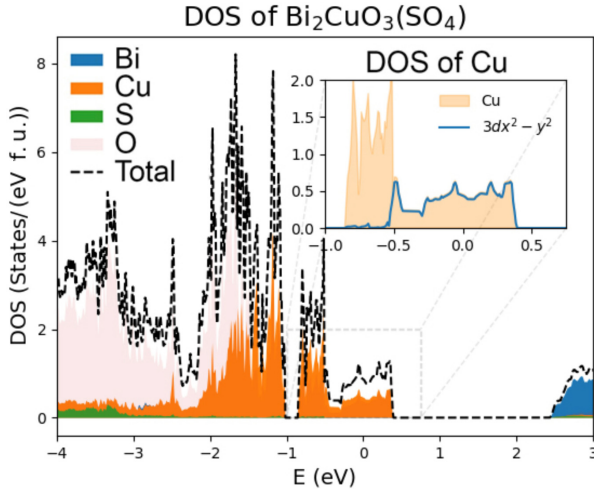


FIG. 5. Calculated electronic density of states for $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ at the PBE level. Colored areas represent the element-resolved contributions of Bi, Cu, S, and O states, while the dashed line shows the total DOS. The Fermi level is set to $E = 0$ eV. The states in the vicinity of the Fermi level are dominated by Cu $3d$ contributions. Inset: Cu-projected DOS highlighting the dominant contribution of the $3d_{x^2-y^2}$ orbital.

parameters $A_1 = 6.59$, $\theta_{D_1} = 946.6$ K, $A_2 = 4.41$, $\theta_{D_2} = 188.5$ K. While this fit does not account for the magnetic contribution, it highlights deviations from the simple Debye behavior. They are especially pronounced at 15 – 18 K where a kink is visible in C_p/T . This kink is reproducible between the different batches [24]. It matches the temperature of the susceptibility anomaly and confirms its bulk nature. We thus conclude that $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ undergoes magnetic ordering below 16 K.

F. Microscopic magnetic model

PBE calculations for the band structure of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ are fully consistent with expectations for an insulating Cu^{2+} -based compound [32,43–45] (Fig. 5). The valence band is predominantly composed of Cu $3d$ and O $2p$ states. The states above -0.6 eV are dominated by the Cu $3d_{x^2-y^2}$ orbital, in line with the anticipated ligand-field splitting. Unoccupied states above 2 eV arise from Bi $6p$ orbitals. The partial densities of states clearly identify Cu^{2+} ions as the magnetic ions of the material, and Cu $3d$ bands additionally form narrow features close to the Fermi level. The metallic PBE spectrum contradicts the experimentally observed insulating behavior inferred from the green color of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$. This discrepancy stems from strong correlation effects within the partially filled Cu $3d$ shell that PBE significantly underestimates. These missing correlations can be incorporated either on the level of an effective model or within the mean-field DFT + U approach.

We focus on the narrow electronic bands intersecting the Fermi level. The bands are characterized by a dominant Cu $3d_{x^2-y^2}$ orbital contribution, with the local orbital axes aligned along the shorter Cu–O bonds. In $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$, the Cu^{2+} ions are situated in a strongly elongated octahedral coordination CuO_{4+2} , comprising four short

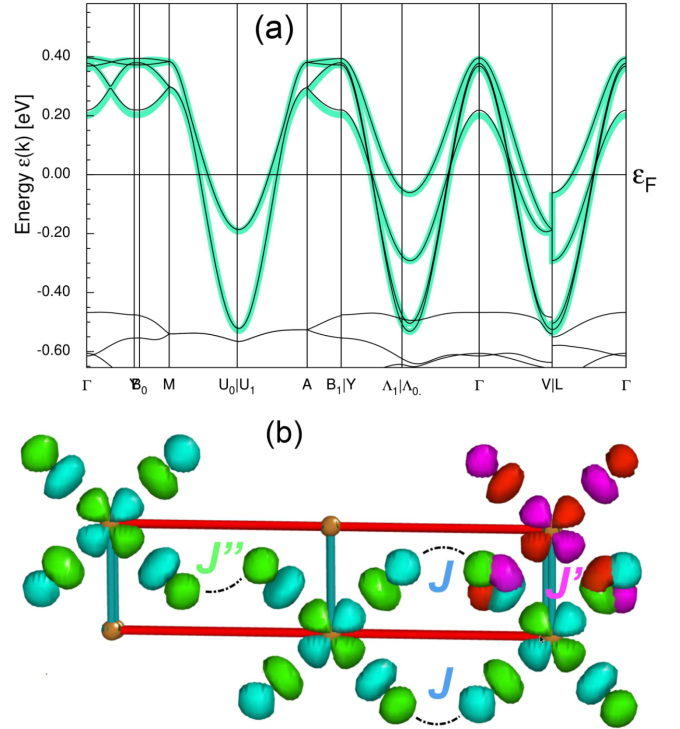


FIG. 6. (a) PBE band structure (thin black lines) and the fit with the tight-binding model (thick green lines). (b) Magnetic orbitals represented through Wannier functions, where the Cu $3d_{x^2-y^2}$ orbital exhibits pronounced hybridization with the surrounding O $2p$ states. Oxygen atoms are omitted for clarity.

in-plane Cu–O bonds (1.96–1.98 Å) and two significantly longer apical bonds (2.2 Å and 2.79 Å). This pronounced anisotropy of the local ligand environment places the $3d_{x^2-y^2}$ orbital highest in energy within the crystal-field scheme, in full agreement with the PBE results. The $3d_{x^2-y^2}$ -derived bands were subsequently described with an effective tight-binding model by constructing Wannier functions centered on the Cu sites [31]. The resulting Wannier fit accurately reproduces the DFT band dispersion and allows for the extraction of the relevant Cu–Cu hopping integrals t_i (Fig. 6 and Table II).

TABLE II. Cu–Cu distances (in Å), hopping parameters t_i (in meV), and exchange couplings J_i (in K) in $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$. The AFM contributions J_i^{AFM} are calculated as $4t_i^2/U_{\text{eff}}$ with $U_{\text{eff}} = 8.5$ eV. The total exchange couplings J_i are obtained from DFT + U calculations ($U_{\text{Cu}} = 8.5$ eV, $J_{\text{Cu}} = 1$ eV). The notation of J_i is illustrated in Fig. 1. The structural data reported in Ref. [22] were used as input for the DFT calculations.

Coupling	$d_{\text{Cu-Cu}}$	t_i	J_i^{AFM}	J_i
Rung J'	2.79	61	20.2	−133.5
Leg J	5.40	172	161.6	194.7
Diag J''	6.07	−51	14.3	6.2
$J_{\perp 1}$	5.68	7	0.3	−2.8
$J_{\perp 2}$	6.09	−40	8.9	6.1
$J_{\parallel}$	8.20	−0.2	3×10^{-4}	1.9
	8.49	−24	3.1	1.9

Additionally, we estimate the exchange couplings using the DFT + U mapping approach that yields a sum of FM and AFM contributions. Comparison between J_i^{AFM} and the total exchange couplings J_i obtained from DFT + U calculations provides direct insight into the competition between AFM and FM exchange mechanisms. In particular, the rung coupling J' exhibits only a modest AFM contribution from the hopping analysis, whereas the total exchange becomes strongly ferromagnetic due to sizable FM contributions beyond the one-orbital picture. In contrast, the leg coupling (J) remains predominantly antiferromagnetic because of the efficient Cu–O–Cu superexchange pathway revealed by the Wannier functions.

The real-space Wannier functions are shown in Fig. 6(b) and reveal a magnetic orbital of predominant Cu $3d_{x^2-y^2}$ character, strongly hybridized with the surrounding O $2p$ states that play a key role in mediating magnetic interactions. The Wannier representation provides direct insight into the exchange pathways along the spin ladder. The rung coupling J' is governed by a Cu–O–Cu superexchange geometry with an angle of $90.11(1)^\circ$, which facilitates a FM interaction. In contrast, both the leg coupling J and the diagonal coupling J'' are AFM and originate from extended Cu–O–O–Cu superexchange paths involving orbitals of several oxygen atoms. The drastic difference between J and J'' is surprising in view of the same O–O contact and the very similar Cu–O–O angles of about 134° . An important geometrical difference is that the relevant p orbitals of the oxygen atoms overlap in the case of J , whereas they are offset in the case of J'' , thus hindering electron hopping and superexchange (Fig. 6).

Based on the calculated exchange parameters summarized in Table II, we conclude that the spin lattice of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ comprises nonfrustrated spin ladders with FM rungs and AFM legs. The diagonal coupling J'' is smaller than J by more than one order of magnitude and, therefore, negligible. The interladder couplings can be classified into two distinct types according to their geometry. The couplings $J_{\perp 1}$ and $J_{\perp 2}$ connect spins located on opposite legs of the neighboring ladders. In contrast, the coupling $J_{\parallel}$ links spins belonging to the same legs. The interladder interactions $J_{\perp 1}$, $J_{\perp 2}$, and $J_{\parallel}$ are significantly weaker than J and J' . Therefore, $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ should be deemed a quasi-1D magnet.

G. Comparison to the experiment

To facilitate a quantitative comparison between the exchange parameters obtained from DFT and the experimental data, we perform QMC simulations of the magnetic susceptibility. The inclusion of a weak diagonal interaction does not significantly affect the thermodynamic response of the ladder system [46]. Therefore, only the dominant rung and leg couplings were retained in the simulations. Since QMC simulations yield dimensionless quantities, the computed observables were rescaled using appropriate normalization factors to enable a direct comparison with the experimental results. To this end, dimensionless forms of the susceptibility, spin gap, and temperature were introduced:

$$\chi^* = \frac{\chi |J'|}{N_A (g \mu_B)^2}, \quad \Delta^* = \frac{\Delta}{|J'|}, \quad t = \frac{k_B T}{|J'|}. \quad (7)$$

All quantities were expressed in units of a characteristic energy scale, which was chosen as the absolute value of the rung coupling $|J'|$. Fixing this scale to unity, a series of simulations was performed for different ratios $J/|J'|$, allowing us to systematically explore the influence of the leg coupling strength.

Preliminary QMC simulations performed for the coupling ratios in the range of $1.0 \leq J/|J'| \leq 2.5$ reveal simple empirical linear relations between the ratios $J/|J'|$, $|J'|/J$, and the position of the susceptibility maximum, T_{max} :

$$\begin{aligned} \frac{J}{|J'|} &= \frac{T_{\text{max}}}{|J'|} m_1 + b_1, \\ \frac{|J'|}{J} &= \frac{T_{\text{max}}}{J} m_2 + b_2, \end{aligned} \quad \Leftrightarrow \begin{pmatrix} 1 & -b_1 \\ -b_2 & 1 \end{pmatrix} \begin{pmatrix} J \\ |J'| \end{pmatrix} = T_{\text{max}} \begin{pmatrix} m_1 \\ m_2 \end{pmatrix},$$

where $m_1 = 1.553$, $b_1 = -0.057$ and $m_2 = -0.582$, $b_2 = 1.294$ are the corresponding linear regression parameters. The resulting system of equations enables a straightforward estimation of both exchange constants using only the experimentally determined position of the susceptibility maximum. From $T_{\text{max}} \approx 156$ K, the exchange couplings are estimated as $J \approx 231$ K and $|J'| \approx 208$ K, yielding $J/|J'| \approx 1.24$. Since these values are subject to uncertainty inherent to the empirical parametrization, they were employed as initial estimates for the subsequent simulations of the full susceptibility curve.

The results obtained for 31 discrete values of the anisotropy ratios $J/|J'|$ within the interval $[1.0, 1.31]$ were subsequently fitted using Padé approximants following the procedure described in Ref. [39]. This approach enables a smooth interpolation of the ratio $J/|J'|$ and allows for a continuous fitting of the experimental data. The experimental magnetic susceptibility was then modeled by introducing four additional free parameters, χ_0 , C_{imp} , θ_{imp} , and Landé g -factor, according to

$$\chi_m = \chi_0 + \frac{C_{\text{imp}}}{T - \theta_{\text{imp}}} + \frac{N_A g^2 \mu_B^2}{|J'|} \chi^*. \quad (8)$$

The best fit of the experimental magnetic susceptibility in the whole temperature range returns $J = 258$ K, $J' = -208$ K, $g = 2.14$ together with $\chi_0 = -0.35 \times 10^{-4}$ emu mol $^{-1}$, $C_{\text{imp}} = 9.10 \times 10^{-3}$ emu K mol $^{-1}$, and $\theta_{\text{imp}} = -7$ K. The interaction parameter of the impurity part is consistent with the analysis of the ESR signal that returned $\theta_{\text{imp}} = -10$ K (Sec. III D), whereas the C_{imp} value corresponds to 2.4% of the impurity spins.

The spin gap was extracted for each simulated susceptibility curve by fitting the low-temperature data using the expression proposed in Ref. [47]: $\chi^* = (A/\sqrt{t}) e^{-\Delta^*/t}$, where $\Delta^* = \Delta/|J'|$. The resulting Δ^* values listed in Table S1 [24] were subsequently rescaled by the magnitude of the rung coupling $|J'|$. The spin gaps obtained for the investigated coupling ratios amount to approximately 10% of the rung exchange interaction J' . Using the exchange parameters from the susceptibility fit, we find $\Delta \simeq 28$ K for the isolated spin ladder in $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$.

IV. DISCUSSION AND SUMMARY

The magnetic properties of $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ are successfully rationalized within a two-leg spin-ladder model. The diagonal

TABLE III. Comparison of the leg (J_{leg}) and rung (J_{rung}) exchange couplings in representative two-leg $S = \frac{1}{2}$ spin-ladder compounds. For each material, the dominant superexchange pathways (S for short-range and L for long-range) and the sign of the interactions (AFM or FM) are indicated. Exchange constants are given in Kelvin; negative values denote ferromagnetic couplings. For $(\text{CPA})_2\text{CuBr}_4$, the exchange couplings are averaged over two inequivalent ladders reported in Ref. [51]. Experimental values (fits to the magnetic susceptibility and/or excitation spectra) are given in each case.

Compound	J_{rung} (K)	J_{leg} (K)	Reference(s)
BPCB	13 (L)	3 (L)	[52,53]
DLCB	7 (L)	7 (L)	[54]
$(\text{CPA})_2\text{CuBr}_4$	10 (L)	9 (L)	[51]
IPA-CuCl ₃	−27 (S)	14 (L)	[55]
PHCC	23 (S)	14 (L)	[56]
DIMPY	9 (L)	17 (L)	[57,58]
Ba ₂ CuTeO ₆	~90 (L)	~90 (L)	[8,49]
BiCu ₂ PO ₆	~110 (L)	~110 (L)	[16,19]
KCu ₂ BiO ₂ (SO ₄) ₂	205 (L)	117 (L)	[59]
Bi ₂ CuO ₃ (SO ₄)	−208 (S)	258 (L)	this work

couplings within the ladder are very weak, leading to only a minor frustration. An interesting feature of the material is its combination of the FM rung coupling and AFM leg coupling of comparable strength, in contrast to the majority of Cu-based spin ladders, which are purely AFM. The FM nature of the rung coupling leads to a rather small spin gap, less than 10% of J . Therefore, materials with the FM-AFM spin ladders are more prone to developing long-range magnetic order even from weak interladder couplings. We have verified this by performing simulations for coupled spin ladders with $J/|J'| = 1.24$, as determined experimentally, and observed the transition into the long-range-ordered state at $J_{\perp 2}/|J'| = 0.016$ [24], which is about half of the DFT estimate of 0.029. It is then natural to attribute the weak magnetic susceptibility and specific heat anomalies at $T_N = 16$ K to the onset of long-range magnetic order. With T_N/J as low as 0.06, most of the magnetic entropy is released above T_N , and the transition manifests itself only by a small kink in C_p/T instead of a larger λ -type anomaly. This situation resembles some other spin-ladder compounds, such as Ba₂CuTeO₆ where magnetic ordering leads to a tiny anomaly in the magnetic susceptibility with no clear signatures in the specific heat [48,49].

It is further instructive to compare the coupling strength in various Cu-based spin-ladder compounds. Their rung couplings can be either short-range (mediated by a Cu-L-Cu bridge, where L is the ligand atom) or long-range (mediated by several ligand atoms), whereas leg couplings are typically long-range. Relevant materials and their exchange parameters are summarized in Table III. One sees that FM rungs are rare and only occur in geometries with the short-range couplings. Even in this case, represented by the Cu₂O₆ dimers, the rung coupling can be either weakly FM or weakly AFM, as exemplified by IPA-CuCl₃ and PHCC, respectively. With

its Cu–O–Cu angle of 90.1°, Bi₂CuO₃(SO₄) stands out as the spin-ladder material with a strong FM rung exchange. The magnitude of the FM exchange in this case is comparable to materials where CuO₄ plaquettes condense into chains with the strong FM nearest-neighbor coupling, such as $J_1 = -228$ K in Li₂CuO₂ [50].

The leg coupling in Bi₂CuO₃(SO₄) is equally remarkable. It largely exceeds the respective couplings in other spin-ladder magnets with long-range superexchange. In fact, $J = 258$ K manifests one of the strongest long-range superexchange couplings among all Cu²⁺ materials reported to date. Whereas Cu–O–O–Cu pathways are known to be efficient because of the large O $2p$ contribution to the respective Wannier functions (Fig. 6), the resulting exchange couplings rarely exceed 100 K, given the large separation of at least 5 Å between the interacting Cu²⁺ ions. The J value in Bi₂CuO₃(SO₄) lies well beyond this typical range. A similarly strong coupling of about 260 K has been reported in Na₂Cu₂TeO₆ [60,61]. In both cases, the CuO₄ plaquettes are perfectly co-aligned, thanks to the bonding network created by Bi³⁺ and Te⁶⁺ forming short contacts to oxygen. While these observer atoms do not contribute any states near the Fermi level and do not directly participate in superexchange, they apparently play an important role in stabilizing the optimal geometry for the long-range superexchange coupling.

In summary, we reported magnetic properties and determined microscopic interaction parameters in the spin-ladder material Bi₂CuO₃(SO₄). The established spin-ladder model features ferromagnetic rungs and antiferromagnetic legs. Both couplings largely exceed in magnitude magnetic interaction parameters in other spin-ladder materials with similar superexchange geometries. A weak anomaly is consistently observed around 16 K and indicates a likely transition into a magnetically ordered state stabilized by non-negligible interladder couplings. A direct microscopic determination of the magnetic structure, however, requires further studies using local or diffraction-based magnetic probes.

ACKNOWLEDGMENTS

Computations for this work were done (in part) using resources of the Leipzig University Computing Center. The work in Dresden was supported by the DFG through the Würzburg-Dresden Cluster of Excellence on Complexity, Topology and Dynamics in Quantum Matter—ctd.qmat (EXC 2147, Project No. 390858490) and the SFB 1143 (Project No. 247310070), as well as by HLD at HZDR, member of the European Magnetic Field Laboratory (EMFL). V.A.G. acknowledges the personal scholarship from DAAD under Grant No. 91837667. We acknowledge ESRF for providing the beamtime at ID22 and thank A. Fitch, C. Dejoie, and J. Gainza for their technical support during the measurement.

DATA AVAILABILITY

The data that support the findings of this article are openly available [24,62].

- [1] F. D. M. Haldane, Continuum dynamics of the 1-D Heisenberg antiferromagnet: Identification with the O(3) nonlinear sigma model, *Phys. Lett. A* **93**, 464 (1983).
- [2] E. Dagotto and T. M. Rice, Surprises on the way from one- to two-dimensional quantum magnets: The ladder materials, *Science* **271**, 618 (1996).
- [3] D. S. Inosov, Quantum magnetism in minerals, *Adv. Phys.* **67**, 149 (2018).
- [4] H. Zhang, Z. Zhao, D. Gautreau, M. Raczkowski, A. Saha, V. O. Garlea, H. Cao, T. Hong, H. O. Jeschke, S. D. Mahanti, T. Birol, F. F. Assaad, and X. Ke, Coexistence and interaction of spinons and magnons in an antiferromagnet with alternating antiferromagnetic and ferromagnetic quantum spin chains, *Phys. Rev. Lett.* **125**, 037204 (2020).
- [5] P. Lemmens, G. Güntherodt, and C. Gros, Magnetic light scattering in low-dimensional quantum spin systems, *Phys. Rep.* **375**, 1 (2003).
- [6] S. Lebernegg, O. Janson, I. Rousochatzakis, S. Nishimoto, H. Rosner, and A. A. Tsirlin, Frustrated spin chain physics near the Majumdar-Ghosh point in szenicsite $\text{Cu}_3(\text{MoO}_4)(\text{OH})_4$, *Phys. Rev. B* **95**, 035145 (2017).
- [7] A. A. Kulbakov, D. Y. Kononenko, S. Nishimoto, Q. Stahl, A. M. Chakkingal, M. Feig, R. Gumenuik, Y. Skourski, L. Bhaskaran, S. A. Zvyagin, J. P. Embs, I. Puente-Orench, A. Wildes, J. Geck, O. Janson, D. S. Inosov, and D. C. Peets, Coupled frustrated ferromagnetic and antiferromagnetic quantum spin chains in the quasi-one-dimensional mineral antlerite $\text{Cu}_3\text{SO}_4(\text{OH})_4$, *Phys. Rev. B* **106**, L020405 (2022).
- [8] A. Glamazda, Y. S. Choi, S.-H. Do, S. Lee, P. Lemmens, A. N. Ponomaryov, S. A. Zvyagin, J. Wosnitza, D. P. Sari, I. Watanabe, and K.-Y. Choi, Quantum criticality in the coupled two-leg spin ladder $\text{Ba}_2\text{CuTeO}_6$, *Phys. Rev. B* **95**, 184430 (2017).
- [9] A. A. Tsirlin, I. Rousochatzakis, D. Kasinathan, O. Janson, R. Nath, F. Weickert, C. Geibel, A. M. Läuchli, and H. Rosner, Bridging frustrated-spin-chain and spin-ladder physics: Quasi-one-dimensional magnetism of BiCu_2PO_6 , *Phys. Rev. B* **82**, 144426 (2010).
- [10] M. Azuma, Z. Hiroi, M. Takano, K. Ishida, and Y. Kitaoka, Observation of a spin gap in SrCu_2O_3 comprising spin- $\frac{1}{2}$ quasi-1D two-leg ladders, *Phys. Rev. Lett.* **73**, 3463 (1994).
- [11] V. Kiryukhin, Y. J. Kim, K. J. Thomas, F. C. Chou, R. W. Erwin, Q. Huang, M. A. Kastner, and R. J. Birgeneau, Magnetic properties of the $S = \frac{1}{2}$ quasi-one-dimensional antiferromagnet CaCu_2O_3 , *Phys. Rev. B* **63**, 144418 (2001).
- [12] S. Sugai, T. Shinoda, N. Kobayashi, Z. Hiroi, and M. Takano, Anisotropic exchange integrals in the two-leg spin ladder $\text{LaCuO}_{2.5}$, *Phys. Rev. B* **60**, R6969(R) (1999).
- [13] B. Koteswararao, S. Salunke, A. V. Mahajan, I. Dasgupta, and J. Bobroff, Spin-gap behavior in the two-leg spin-ladder BiCu_2PO_6 , *Phys. Rev. B* **76**, 052402 (2007).
- [14] O. Mentr , E. Janod, P. Rabu, M. Hennion, F. Leclercq-Hugueux, J. Kang, C. Lee, M.-H. Whangbo, and S. Petit, Incommensurate spin correlation driven by frustration in BiCu_2PO_6 , *Phys. Rev. B* **80**, 180413(R) (2009).
- [15] K.-Y. Choi, J. W. Hwang, P. Lemmens, D. Wulferding, G. J. Shu, and F. C. Chou, Evidence for dimer crystal melting in the frustrated spin-ladder system BiCu_2PO_6 , *Phys. Rev. Lett.* **110**, 117204 (2013).
- [16] K. W. Plumb, K. Hwang, Y. Qiu, L. W. Harriger, G. E. Granroth, A. I. Kolesnikov, G. J. Shu, F. C. Chou, Ch. R egg, Y. B. Kim, and Y.-J. Kim, Quasiparticle-continuum level repulsion in a quantum magnet, *Nat. Phys.* **12**, 224 (2016).
- [17] Y. Kohama, S. Wang, A. Uchida, K. Prsa, S. Zvyagin, Y. Skourski, R. D. McDonald, L. Balicas, H. M. R nnow, C. R egg, and M. Jaime, Anisotropic cascade of field-induced phase transitions in the frustrated spin-ladder system BiCu_2PO_6 , *Phys. Rev. Lett.* **109**, 167204 (2012).
- [18] F. Casola, T. Shiroka, A. Feiguin, S. Wang, M. S. Grbi , M. Horvati , S. Kr mer, S. Mukhopadhyay, K. Conder, C. Berthier, H.-R. Ott, H. M. R nnow, Ch. R egg, and J. Mesot, Field-induced quantum soliton lattice in a frustrated two-leg spin-1/2 ladder, *Phys. Rev. Lett.* **110**, 187201 (2013).
- [19] L. Splinter, N. A. Drescher, H. Krull, and G. S. Uhrig, Minimal model for the frustrated spin ladder system BiCu_2PO_6 , *Phys. Rev. B* **94**, 155115 (2016).
- [20] K. Hwang and Y. B. Kim, Theory of triplon dynamics in the quantum magnet BiCu_2PO_6 , *Phys. Rev. B* **93**, 235130 (2016).
- [21] P. Pilch, K. Amelin, G. Schmiedinghoff, A. Reinold, C. Zhu, K. Yu. Povarov, S. Zvyagin, H. Engelkamp, Y.-P. Lan, G.-J. Shu, F. C. Chou, U. Nagel, T. R  m, G. S. Uhrig, B. Fauseweh, and Z. Wang, Low-energy spin excitations in field-induced phases of the spin-ladder antiferromagnet BiCu_2PO_6 , *Phys. Rev. B* **111**, 024423 (2025).
- [22] O. Sidra, D. Charkin, I. Plokhikh, E. Nazarchuk, A. Holzheid, and G. Akimov, Expanding family of litharge-derived sulfate minerals and synthetic compounds: Preparation and crystal structures of $[\text{Bi}_2\text{CuO}_3]\text{SO}_4$ and $[\text{Ln}_2\text{O}_2]\text{SO}_4$ ($\text{Ln} = \text{Dy}$ and Ho), *Minerals* **10**, 887 (2020).
- [23] M. L , M. Colmont, H. Kabbour, S. Colis, and O. Mentr , Revised Bi/M layered oxo-sulfate ($\text{M} = \text{Co}, \text{Cu}$): A structural and magnetic study, *Inorg. Chem.* **53**, 6969 (2014).
- [24] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/nwlh-vnyp> for additional details of the crystal structure, comparison between the different batches, and for the simulated magnetization curves.
- [25] A. Fitch, C. Dejoie, E. Covacci, G. Confalonieri, O. Grendal, L. Claustre, P. Guillou, J. Kieffer, W. de Nolf, S. Petitdemange, M. Ruat, and Y. Watier, ID22—the high-resolution powder-diffraction beamline at ESRF, *J. Synchrotron Rad.* **30**, 1003 (2023).
- [26] V. Pet   ek, M. Du ek, and L. Palatinus, Crystallographic computing system JANA2006: General features, *Z. Krist.* **229**, 345 (2014).
- [27] B. Aurivillius, Pyrolysis products of $\text{Bi}_2(\text{SO}_4)_3$. Crystal structures of $\text{Bi}_{26}\text{O}_{27}(\text{SO}_4)_{12}$ and $\text{Bi}_{14}\text{O}_{16}(\text{SO}_4)_5$, *Acta Chem. Scand.* **41a**, 415 (1987).
- [28] S. A. Zvyagin, J. Krzystek, P. H. M. van Loosdrecht, G. Dhalenne, and A. Revcolevschi, High-field ESR study of the dimerized-incommensurate phase transition in the spin-Peierls compound CuGeO_3 , *Physica B* **346-347**, 1 (2004).
- [29] K. Koepernik and H. Eschrig, Full-potential nonorthogonal local-orbital minimum-basis band-structure scheme, *Phys. Rev. B* **59**, 1743 (1999).
- [30] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized gradient approximation made simple, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [31] H. Eschrig and K. Koepernik, Tight-binding models for the iron-based superconductors, *Phys. Rev. B* **80**, 104503 (2009).

- [32] V. V. Mazurenko, S. L. Skornyakov, A. V. Kozhevnikov, F. Mila, and V. I. Anisimov, Wannier functions and exchange integrals: The example of LiCu_2O_2 , *Phys. Rev. B* **75**, 224408 (2007).
- [33] V. V. Mazurenko, M. V. Valentyuk, R. Stern, and A. A. Tsirlin, Nonfrustrated interlayer order and its relevance to the Bose-Einstein condensation of magnons in $\text{BaCuSi}_2\text{O}_6$, *Phys. Rev. Lett.* **112**, 107202 (2014).
- [34] P. Bag, N. Ahmed, V. Singh, M. Sahoo, A. A. Tsirlin, and R. Nath, Low-dimensional magnetism of $\text{BaCuTe}_2\text{O}_6$, *Phys. Rev. B* **103**, 134410 (2021).
- [35] H. J. Xiang, E. J. Kan, S.-H. Wei, M.-H. Whangbo, and X. G. Gong, Predicting the spin-lattice order of frustrated systems from first principles, *Phys. Rev. B* **84**, 224429 (2011).
- [36] S. Todo and K. Kato, Cluster algorithms for general- S quantum spin systems, *Phys. Rev. Lett.* **87**, 047203 (2001).
- [37] A. F. Albuquerque, F. Alet, P. Corboz, P. Dayal, A. Feiguin, S. Fuchs, L. Gamper, E. Gull, S. Gürtler, A. Honecker, R. Igarashi, M. Körner, A. Kozhevnikov, A. Läuchli, S. Manmana, M. Matsumoto, I. P. McCulloch, F. Michel, R. M. Noack, G. Pawłowski, *et al.*, The ALPS project release 1.3: Open-source software for strongly correlated systems, *J. Magn. Magn. Mater.* **310**, 1187 (2007).
- [38] A. F. Albuquerque, M. Troyer, and J. Oitmaa, Quantum phase transition in a Heisenberg antiferromagnet on a square lattice with strong plaquette interactions, *Phys. Rev. B* **78**, 132402 (2008).
- [39] D. C. Johnston, R. K. Kremer, M. Troyer, X. Wang, A. Klümper, S. L. Bud'ko, A. F. Panchula, and P. C. Canfield, Thermodynamics of spin $S = 1/2$ antiferromagnetic uniform and alternating-exchange Heisenberg chains, *Phys. Rev. B* **61**, 9558 (2000).
- [40] M. Sigrist and A. Furusaki, Low-temperature properties of the randomly depleted Heisenberg ladder, *J. Phys. Soc. Jpn.* **65**, 2385 (1996).
- [41] H.-J. Mikeska, U. Neugebauer, and U. Schollwöck, Spin ladders with nonmagnetic impurities, *Phys. Rev. B* **55**, 2955 (1997).
- [42] D. Schmidiger, K. Yu. Povarov, S. Galeski, N. Reynolds, R. Bewley, T. Guidi, J. Ollivier, and A. Zheludev, Emergent interacting spin islands in a depleted strong-leg Heisenberg ladder, *Phys. Rev. Lett.* **116**, 257203 (2016).
- [43] A. A. Tsirlin, O. Janson, and H. Rosner, $\beta\text{-Cu}_2\text{V}_2\text{O}_7$: A spin- $\frac{1}{2}$ honeycomb lattice system, *Phys. Rev. B* **82**, 144416 (2010).
- [44] A. A. Tsirlin and H. Rosner, Uniform spin-chain physics arising from N—C—N bridges in CuNCN , the nitride analog of the copper oxides, *Phys. Rev. B* **81**, 024424 (2010).
- [45] A. A. Tsirlin, R. Zinke, J. Richter, and H. Rosner, Spiral ground state in the quasi-two-dimensional spin- $\frac{1}{2}$ system Cu_2GeO_4 , *Phys. Rev. B* **83**, 104415 (2011).
- [46] D. C. Johnston, M. Troyer, S. Miyahara, D. Lidsky, K. Ueda, M. Azuma, Z. Hiroi, M. Takano, M. Isobe, Y. Ueda, M. A. Korotin, V. I. Anisimov, A. V. Mahajan, and L. L. Miller, Magnetic susceptibilities of spin- $1/2$ antiferromagnetic Heisenberg ladders and applications to ladder oxide compounds, [arXiv:cond-mat/0001147](https://arxiv.org/abs/cond-mat/0001147).
- [47] M. Troyer, H. Tsunetsugu, and D. Würtz, Thermodynamics and spin gap of the Heisenberg ladder calculated by the look-ahead Lanczos algorithm, *Phys. Rev. B* **50**, 13515 (1994).
- [48] G. N. Rao, R. Sankar, A. Singh, I. P. Muthuselvam, W. T. Chen, V. N. Singh, G.-Y. Guo, and F. C. Chou, Tellurium-bridged two-leg spin ladder in $\text{Ba}_2\text{CuTeO}_6$, *Phys. Rev. B* **93**, 104401 (2016).
- [49] A. S. Gibbs, A. Yamamoto, A. N. Yaresko, K. S. Knight, H. Yasuoka, M. Majumder, M. Baenitz, P. J. Saines, J. R. Hester, D. Hashizume, A. Kondo, K. Kindo, and H. Takagi, $S = \frac{1}{2}$ quantum critical spin ladders produced by orbital ordering in $\text{Ba}_2\text{CuTeO}_6$, *Phys. Rev. B* **95**, 104428 (2017).
- [50] W. E. A. Lorenz, R. O. Kuzian, S.-L. Drechsler, W.-D. Stein, N. Wizen, G. Behr, J. Málek, U. Nitzsche, H. Rosner, A. Hiess, W. Schmidt, R. Klingeler, M. Loewenhaupt, and B. Büchner, Highly dispersive spin excitations in the chain cuprate Li_2CuO_2 , *Europhys. Lett.* **88**, 37002 (2009).
- [51] J. Philippe, F. Elson, T. Arh, S. Sanz, M. Metzelaars, D. W. Tam, O. K. Forslund, O. Shliakhtun, C. Jiang, J. Lass, M. D. Le, J. Ollivier, P. Bouillot, T. Giamarchi, M. Bartkowiak, D. G. Mazzone, P. Kögerler, M. Månsson, A. M. Läuchli, Y. Sassa, *et al.*, Magnetic and phononic dynamics in the two-ladder quantum magnet $(\text{C}_5\text{H}_9\text{NH}_3)_2\text{CuBr}_4$, *Phys. Rev. B* **113**, 134438 (2026).
- [52] B. C. Watson, V. N. Kotov, M. W. Meisel, D. W. Hall, G. E. Granroth, W. T. Montfrooij, S. E. Nagler, D. A. Jensen, R. Backov, M. A. Petruska, G. E. Fanucci, and D. R. Talham, Magnetic spin ladder $(\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4$: High-field magnetization and scaling near quantum criticality, *Phys. Rev. Lett.* **86**, 5168 (2001).
- [53] A. T. Savici, G. E. Granroth, C. L. Broholm, D. M. Pajerowski, C. M. Brown, D. R. Talham, M. W. Meisel, K. P. Schmidt, G. S. Uhrig, and S. E. Nagler, Neutron scattering evidence for isolated spin- $\frac{1}{2}$ ladders in $(\text{C}_5\text{D}_{12}\text{N})_2\text{CuBr}_4$, *Phys. Rev. B* **80**, 094411 (2009).
- [54] T. Hong, K. P. Schmidt, K. Coester, F. F. Awwadi, M. M. Turnbull, Y. Qiu, J. A. Rodriguez-Rivera, M. Zhu, X. Ke, C. P. Aoyama, Y. Takano, H. Cao, W. Tian, J. Ma, R. Custelcean, H. D. Zhou, and M. Matsuda, Magnetic ordering induced by interladder coupling in the spin- $\frac{1}{2}$ Heisenberg two-leg ladder antiferromagnet $\text{C}_9\text{H}_{18}\text{N}_2\text{CuBr}_4$, *Phys. Rev. B* **89**, 174432 (2014).
- [55] T. Fischer, S. Duffe, and G. S. Uhrig, Microscopic model for Bose-Einstein condensation and quasiparticle decay, *Europhys. Lett.* **96**, 47001 (2011).
- [56] A. A. Tsirlin, O. Janson, and I. Rousochatzakis, One-dimensional physics of the frustrated quantum magnet PHCC, *Z. Naturforsch. A* **81**, 335 (2026).
- [57] T. Hong, Y. H. Kim, C. Hotta, Y. Takano, G. Tremelling, M. M. Turnbull, C. P. Landee, H.-J. Kang, N. B. Christensen, K. Lefmann, K. P. Schmidt, G. S. Uhrig, and C. Broholm, Field-induced Tomonaga-Luttinger liquid phase of a two-leg spin- $1/2$ ladder with strong leg interactions, *Phys. Rev. Lett.* **105**, 137207 (2010).
- [58] D. Schmidiger, P. Bouillot, S. Mühlbauer, S. Gvasaliya, C. Kollath, T. Giamarchi, and A. Zheludev, Spectral and thermodynamic properties of a strong-leg quantum spin ladder, *Phys. Rev. Lett.* **108**, 167201 (2012).
- [59] L. V. Shvanskaya, T. D. Bushneva, D. A. Chareev, P. A. Maksimov, A. A. Sudakov, E. I. Kornienko, A. V. Ushakov, S. V. Streltsov, and A. N. Vasiliev, Peculiar crystal structure and strong-rung spin ladders in $\text{KCu}_2\text{BiO}_2(\text{SO}_4)_2$, *Inorg. Chem.* **65**, 395 (2026).

- [60] S. Gao, L.-F. Lin, A. F. May, B. K. Rai, Q. Zhang, E. Dagotto, A. D. Christianson, and M. B. Stone, Weakly coupled alternating $S = \frac{1}{2}$ chains in the distorted honeycomb lattice compound $\text{Na}_2\text{Cu}_2\text{TeO}_6$, *Phys. Rev. B* **102**, 220402(R) (2020).
- [61] Y. Shangguan, S. Bao, Z.-Y. Dong, Z. Cai, W. Wang, Z. Huang, Z. Ma, J. Liao, X. Zhao, R. Kajimoto, K. Iida, D. Voneshen, S.-L. Yu, J.-X. Li, and J. Wen, Evidence for strong correlations at finite temperatures in the dimerized magnet $\text{Na}_2\text{Cu}_2\text{TeO}_6$, *Phys. Rev. B* **104**, 224430 (2021).
- [62] R. A. R. Hernandez, K. Yu. Povarov, S. Zvyagin, O. I. Siidra, A. A. Tsirlin, and V. A. Ginga, Supplemental Materials for the manuscript “Quantum spin ladder with ferromagnetic rungs in $\text{Bi}_2\text{CuO}_3(\text{SO}_4)$ ” [Data set], Zenodo, 2026, <https://doi.org/10.5281/zenodo.19241586>.