

Green's function theory of magnetism in Bi_2CuO_4 : Anisotropic Heisenberg XYZ modelR. O. Kuzian^{1,2,*} and E. E. Krasovskii^{1,3,4}¹*Donostia International Physics Center (DIPC), Paseo Manuel de Lardizabal 4, San Sebastián/Donostia, 20018 Basque Country, Spain*²*Frantsevich Institute for Problems of Materials Science, National Academy of Science of Ukraine, 3, str. Omeliana Pritsaka, 03142 Kyiv, Ukraine*³*Departamento de Polímeros y Materiales Avanzados: Física, Química y Tecnología, Universidad del País Vasco-Euskal Herriko Unibertsitatea, Donostia-San Sebastián, 20080 Basque Country, Spain*⁴*IKERBASQUE, Basque Foundation for Science, 48013 Bilbao, Spain*

(Received 23 March 2026; accepted 17 July 2026; published 4 August 2026)

The Green's function theory of Lymar' and Rudoi [Theor. Math. Phys. **21**, 990 (1974)] is generalized to the case of multiple intra- and intersublattice magnetic interactions in a collinear spin-half antiferromagnet and applied to magnetic excitations in Bi_2CuO_4 . The spin Hamiltonian includes both the out-of-plane and in-plane symmetric anisotropy terms and the Zeeman term, which describes the interaction with an external magnetic field B applied along the Néel vector. Within the spin-wave approximation, we calculate spin excitation dispersion, antiferromagnetic resonance frequencies, and the critical field B_c of the spin-flop metamagnetic transition. A weak in-plane anisotropy is shown to result in a gap in the acousticlike branch of the excitations. The gap nonlinearly depends on the external field and closes at $B = B_c$. An expression for the Néel temperature in the Tyablikov random phase approximation at zero field is derived. For the parameters derived from the recent inelastic neutron scattering study by Yuan *et al.* [Phys. Rev. B **103**, 134436 (2021)], it gives $T_N \approx 52$ K.

DOI: [10.1103/bn1b-mt34](https://doi.org/10.1103/bn1b-mt34)

I. INTRODUCTION

Cuprates are a rich family of ternary copper oxides with a plethora of unusual physical properties. The CuO_4 plaquette is a building block of various structures, such as two-dimensional corner-shared CuO_2 planes of high- T_c superconductors [1–6], one-dimensional (1D) corner-shared [7–9], and edge-shared [10–13] chains or peculiarly arranged isolated plaquettes. Close to the Fermi energy, the electronic structure of cuprates is determined by the Cu 3d and oxygen 2p states of the CuO_4 plaquettes. Moreover, the Cu 3d states are so strongly split by the oxygen ligand field that only the $d_{x^2-y^2}$ orbital matters (the axes x and y point from the Cu atom to the nearest oxygen atoms). Thus, a mean-field band structure of cuprates often contains a single band crossing the Fermi level. However, the cuprates with a half-filled band are insulators because of strong many-body correlations. Low-energy electron structure of these compounds is described by effective Hamiltonians, which include both rotationally invariant Heisenberg exchange interactions between the localized spins of the Cu^{2+} ions and anisotropic exchange terms, the single-ion anisotropy terms being prohibited for the spin-half systems. Thus, the insulating cuprates represent a physical

realization of many models of quantum magnetism on lattices of different dimensions and geometries. The Bi_2CuO_4 crystal is a special one because, while having the same chemical formula and ions valences as the high- T_c compounds $R_2\text{CuO}_2$ (R stands for La or a rare-earth ion), it has completely different structure and properties. In the tetragonal crystal structure of Bi_2CuO_4 , the CuO_4 plaquettes are stacked along the c axis, the plaquette plane being perpendicular to the axis [15,16]. The neighboring in-chain plaquettes are twisted with respect to each other by about 33° [16]. The in-chain Cu-Cu distance is much shorter than the interchain one (see Fig. 1). That is why Bi_2CuO_4 was originally considered as a quasi-1D spin-half compound [17,18]. Further studies established that Bi_2CuO_4 is a three-dimensional antiferromagnet with the Néel temperature T_N in the range 42–47 K [16,19–22]. Inelastic neutron scattering (INS) studies [14,23–25] unambiguously evidence that the interchain superexchange interactions are substantially stronger than the intrachain ones. This statement was also supported by the parameter-free density functional theory (DFT) calculation of Ref. [26], which has demonstrated that the naive idea of the interaction decreasing with distance is erroneous.

Even more dramatic is the story of the determination of the ordered moment direction in Bi_2CuO_4 , which seems to be far from finished. The moment orientation parallel to the crystallographic direction $\mathbf{c}$ was proposed in the neutron diffraction studies of Refs. [16,22]. This model was supported by the optical spectroscopy experiments of Refs. [27–29], but it contradicts the magnetization studies of Ref. [21], which show a linear dependence of the magnetization on external magnetic field $\mathbf{B} \parallel \mathbf{c}$ and a metamagnetic spin-flop

*Contact author: r.kuzian@ipms.kyiv.ua

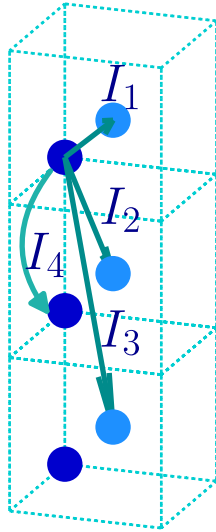


FIG. 1. The unit cell of Bi_2CuO_4 magnetic lattice contains two Cu^{2+} ions, having localized spin-half moments aligned in opposite directions below Néel temperature. Bravais lattice is simple cubic. Schematically shown are the intersite exchange interactions included in the present model following Ref. [14]. The parameters are as follows: $I_1^x = I(1 - \alpha_\perp)$, $I_1^y = I(1 - \alpha_\parallel)$, $I_1^z = I(1 + \alpha_\parallel)$, $I = 4.7$ meV, $\alpha_\perp = 0.013$, $\alpha_\parallel = 4.7 \times 10^{-6}$; the interactions labeled I_{2-4} are assumed to be isotropic with the parameters $I_2 = 1.1$ meV, $I_3 = 0.5$ meV for intersublattice interactions, and $I_4 = 0.36$ meV for a frustrating intrasublattice intrachain interaction.

transition for $\mathbf{B} \perp \mathbf{c}$. Also according to the antiferromagnetic resonance (AFMR) [30–33] and torque magnetometry measurements [34], the moment lies in the $\mathbf{ab}$ plane, $\mathbf{a} \parallel [100]$ and $\mathbf{b} \parallel [010]$. This long-standing controversy was resolved by the polarized neutron scattering measurements of Ref. [35] that definitely showed that the moment lies in the $\mathbf{ab}$ plane. However, the exact direction of the moment in the plane is still under question. The magnetization measurements reported in Ref. [35] indicate that the moment lies either along $\mathbf{a}$ or along $\mathbf{b}$. At the same time, the neutron scattering in the applied field [14] and recent nonreciprocal directional dichroism results [36,37] are interpreted within the assumption that the moment is parallel to either $[110]$ or $[1\bar{1}0]$ direction.

Besides its interest for fundamental physics, the recently discovered magnetically induced ferroelectricity and linear magnetoelectric coupling [35] open a way to control the direction of the spontaneous magnetic moment by electric field, which makes Bi_2CuO_4 a model material to explore the perspective of higher-capacity memories [36,37].

In this paper, we address the magnetism in Bi_2CuO_4 with a two-time temperature Green's functions (GF) method [38–41]. We follow the work of Lymar' and Rudoi (LR) [42], who considered an anisotropic two-sublattice antiferromagnet (XYZ model) in a longitudinal external magnetic field and included only nearest-neighbor nonfrustrated intersublattice exchange interaction. Here, we generalize the LR theory by including multiple inter- and intrasublattice interactions. The latter are frustrating if their exchange parameters have antiferromagnetic sign. The present formalism is an improvement over that of Ref. [26], in which the easy-axis

XXZ model was assumed. We adopt the model proposed in Ref. [14] with four intersite exchange interactions (see Fig. 1) and show that the spectrum of excitations above a collinear antiferromagnetic ground state consists of two gapped branches. The widths of the gaps depend on the longitudinal external magnetic field and are proportional to the AFMR frequencies [30–33]. For a general XYZ model, the field dependence is nonlinear in contrast to the easy-axis XXZ model, where the linear dependence on the field makes the critical field of the spin-flop transition proportional to the gap width at zero field [43]. We generalize Eq. (3.7) of the LR theory [42], which relates the critical field to the smallest of the anisotropy parameters. Thus, we show that the anisotropy parameters may be found from AFMR and magnetization measurements, which are independent of INS ones.

The paper is organized as follows. In Sec. II, we introduce the model and obtain the retarded GF in the Tyablikov random phase approximation (RPA) [39,42,44]. The low temperature spectrum in the spin-wave approximation is analyzed in Sec. III. In Sec. IV, we find the anisotropy parameters and in Sec. V calculate the Néel temperature. The discussion of the results, comparison with the previous works, and conclusions are in Sec. VI. Appendix A contains details of the GF calculation; Appendix B shows the equivalence of the GF approach and linear spin-wave theory (LSWT) at low temperatures. In Appendix C, we compare our theory with that of Ref. [14] in application to a simplified model with $I_2 = I_3 = I_4 = 0$.

II. GREEN'S FUNCTIONS FOR XYZ SPIN HAMILTONIAN

Below, we calculate the retarded GF in the Tyablikov approximation for the XYZ model. We will generalize the consideration of Ref. [42] by taking into account multiple exchange interactions and the specific geometry of Bi_2CuO_4 . The model is given by the anisotropic Heisenberg Hamiltonian

$$\hat{H} = \hat{H}_A + \hat{H}_B + \hat{H}_{AB} + \hat{H}_Z, \quad (1)$$

$$\hat{H}_{A(B)} = \frac{1}{2} \sum_{\mathbf{R} \in A(B)} \sum_{\mathbf{g}, \gamma} I_{\mathbf{g}}^\gamma \hat{S}_{\mathbf{R}}^\gamma \hat{S}_{\mathbf{R}+\mathbf{g}}^\gamma, \quad (2)$$

$$\hat{H}_{AB} = \sum_{\mathbf{R} \in A} \sum_{\mathbf{f}, \gamma} I_{\mathbf{f}}^\gamma \hat{S}_{\mathbf{R}}^\gamma \hat{S}_{\mathbf{R}+\mathbf{f}}^\gamma, \quad (3)$$

$$\hat{H}_Z = -g_z \mu_B B \sum_{\mathbf{R} \in A} (\hat{S}_{\mathbf{R}}^z + \hat{S}_{\mathbf{R}+\rho_B}^z), \quad (4)$$

where $\gamma = x, y$, or z , and $\mathbf{R}$ runs over sites in one sublattice, where the copper spins $S = 1/2$ are located. Vector $\mathbf{g}$ connects interacting sites in the same sublattice and $\mathbf{f}$ between the sublattices. The special feature of the magnetic structure of Bi_2CuO_4 is that the magnetic unit cell is half of the structure unit cell. It contains two sublattices, $\mathbf{R}_A = \mathbf{n} + \rho_A$ and $\mathbf{R}_B = \mathbf{n} + \rho_B$, where $\mathbf{n} = n_1 \mathbf{a} + n_2 \mathbf{b} + n_3 \mathbf{c}/2$ are lattice translations in terms of the basis vectors $\mathbf{a}$, $\mathbf{b}$, and $\mathbf{c}$ of the tetragonal lattice. The positions of the two sites are $\rho_A = 0$ and $\rho_B = \mathbf{a}/2 + \mathbf{b}/2 - 2z_{\text{Cu}} \mathbf{c}$, with $z_{\text{Cu}} = 0.0766$ [16,21] (see Fig. 1). The direction of the spin quantization axis $\hat{z}$ is parallel to the Néel vector $\mathbf{l} \equiv \langle \hat{S}_{\mathbf{R}_A} \rangle - \langle \hat{S}_{\mathbf{R}_B} \rangle$, where $\langle \hat{S}_{\mathbf{R}_{A(B)}} \rangle$ is the spontaneous spin moment proportional to the sublattice

magnetization. As mentioned in the Sec. I, its direction is still not established: According to Ref. [35], $\hat{z} \parallel \mathbf{a}$, while according to Refs. [14,36,37], $\hat{z} \parallel \mathbf{a} + \mathbf{b}$. Our results are the same for both choices of $\hat{z}$. We then choose $\hat{x} = -\hat{c}$ and $\hat{y} = \hat{z} \times \hat{x}$. We follow the model proposed in Ref. [14], which contains a set of four interaction bonds. Only the interaction along one of the bonds, labeled I_1 in Fig. 1, is anisotropic. For $\hat{z} \parallel \mathbf{a} + \mathbf{b}$, our frame coincides with the (x', y', z') frame of Appendix 2 of Ref. [14] for $\phi = 0$, so that $I_1^x = I(1 - \alpha_\perp)$, $I_1^y = I(1 - \alpha_\parallel)$, and $I_1^z = I(1 + \alpha_\parallel)$, and the anisotropy parameters, $\alpha_\perp > \alpha_\parallel > 0$, have the same meaning as in Ref. [14]. For the interaction parameters, we adopt the values reported in Ref. [14]: The intersublattice interaction parameters are $I = 4.7$, $I_2 = 1.1$, and $I_3 = 0.5$ meV, and the parameter of the frustrating intrasublattice intrachain interaction is $I_4 = 0.36$ meV. The anisotropy parameters $\alpha_\perp$ and $\alpha_\parallel$ will be defined in Sec. IV.

The Zeeman Hamiltonian $\hat{H}_Z$ describes the interaction with the external magnetic field applied along $\hat{z}$. Its parameters are the g factor of the Cu^{2+} ion in the $\mathbf{ab}$ plane of Bi_2CuO_4 , $g_z \approx 2.04$, and the Bohr magneton $\mu_B \approx 5.7884 \times 10^{-2}$ meV/T. Taking into account the weak magnetic field $B < B_c \ll k_B T_N / (g_z \mu_B)$ at a low temperature $T \ll T_N$ will allow us to determine in Sec. IV the tiny parameter of the magnetic anisotropy in the $\mathbf{ab}$ plane $\alpha_\parallel$ from the value of the spin-flop field B_c .

Let us introduce the spin-deviation operators

$$\begin{aligned} \hat{S}_{\mathbf{R}}^\pm &= \hat{S}_{\mathbf{R}}^x \pm i\hat{S}_{\mathbf{R}}^y, \quad [\hat{S}_{\mathbf{R}}^+, \hat{S}_{\mathbf{R}'}^-] = 2\hat{S}_{\mathbf{R}}^z \delta_{\mathbf{R}\mathbf{R}'}, \\ [\hat{S}_{\mathbf{R}}^\pm, \hat{S}_{\mathbf{R}'}^\pm] &= \mp \hat{S}_{\mathbf{R}}^\pm \delta_{\mathbf{R}\mathbf{R}'}, \end{aligned}$$

and rewrite the Hamiltonians (2) and (3) as

$$\begin{aligned} \hat{H}_{A(B)} &= \frac{1}{2} \sum_{\mathbf{R} \in A(B)} \sum_{\mathbf{g}} [I_{\mathbf{g}}^z \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{g}}^z \\ &+ \frac{I_{\mathbf{g}}}{2} (\hat{S}_{\mathbf{R}}^+ \hat{S}_{\mathbf{R}+\mathbf{g}}^- + \hat{S}_{\mathbf{R}}^- \hat{S}_{\mathbf{R}+\mathbf{g}}^+) \\ &+ \frac{E_{\mathbf{g}}}{4} (\hat{S}_{\mathbf{R}}^+ \hat{S}_{\mathbf{R}+\mathbf{g}}^+ + \hat{S}_{\mathbf{R}}^- \hat{S}_{\mathbf{R}+\mathbf{g}}^-)], \end{aligned} \quad (5)$$

$$\begin{aligned} \hat{H}_{AB} &= \sum_{\mathbf{R} \in A} \sum_{\mathbf{f}} \left[I_{\mathbf{f}}^z \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{f}}^z + \frac{I_{\mathbf{f}}}{2} (\hat{S}_{\mathbf{R}}^+ \hat{S}_{\mathbf{R}+\mathbf{f}}^- + \hat{S}_{\mathbf{R}}^- \hat{S}_{\mathbf{R}+\mathbf{f}}^+) \right. \\ &\left. + \frac{E_{\mathbf{f}}}{4} (\hat{S}_{\mathbf{R}}^+ \hat{S}_{\mathbf{R}+\mathbf{f}}^+ + \hat{S}_{\mathbf{R}}^- \hat{S}_{\mathbf{R}+\mathbf{f}}^-) \right], \end{aligned} \quad (6)$$

where $I_{\mathbf{g}} = (I_{\mathbf{g}}^x + I_{\mathbf{g}}^y)/2$, $I_{\mathbf{f}} = (I_{\mathbf{f}}^x + I_{\mathbf{f}}^y)/2$, $E_{\mathbf{g}} = I_{\mathbf{g}}^x - I_{\mathbf{g}}^y$, and $E_{\mathbf{f}} = I_{\mathbf{f}}^x - I_{\mathbf{f}}^y$.

We introduce the Fourier transform of the spin operators:

$$\hat{S}_{\mathbf{q}s}^\alpha = \frac{1}{\sqrt{N}} \sum_{\mathbf{n}} e^{i\mathbf{q}(\mathbf{n}+\boldsymbol{\rho}_s)} \hat{S}_{\mathbf{n}+\boldsymbol{\rho}_s}^\alpha, \quad (7)$$

where $s = A$ or B is the sublattice index and $\alpha = +, -, \text{ or } z$; N is the total number of the *unit cells*, which coincides with the number of sites in a sublattice. We will calculate the retarded Green's functions for the spin-deviation operators

$$\begin{aligned} G_{s_1 s_2}^{\alpha\beta}(\mathbf{q}, \omega) &\equiv \langle \langle \hat{S}_{-\mathbf{q}s_1}^\alpha | \hat{S}_{\mathbf{q}s_2}^\beta \rangle \rangle \\ &= -i \int_{t'}^\infty dt e^{i\omega(t-t')} \langle [\hat{S}_{-\mathbf{q}s_1}^\alpha(t), \hat{S}_{\mathbf{q}s_2}^\beta(t')] \rangle, \end{aligned} \quad (8)$$

where $\alpha, \beta = + \text{ or } -$, $[\hat{X}, \hat{Y}]$ is a commutator, and $\langle \dots \rangle$ denotes the thermal average:

$$\langle \hat{X} \rangle = Q^{-1} \text{Tr} [e^{-\beta \hat{H}} \hat{X}], \quad Q = \text{Tr} e^{-\beta \hat{H}}. \quad (9)$$

Here, Tr denotes the trace of an operator and $\beta = (k_B T)^{-1}$ is the inverse temperature. The time dependence of an operator $\hat{X}(t)$ is given by $\hat{X}(t) = e^{it\hat{H}} \hat{X} e^{-it\hat{H}}$. We denote

$$\begin{aligned} G_1(\mathbf{q}, \omega) &\equiv \langle \langle \hat{S}_{-\mathbf{q}A}^+ | \hat{S}_{\mathbf{q}A}^- \rangle \rangle, \\ G_2(\mathbf{q}, \omega) &\equiv \langle \langle \hat{S}_{-\mathbf{q}B}^- | \hat{S}_{\mathbf{q}A}^- \rangle \rangle, \\ G_3(\mathbf{q}, \omega) &\equiv \langle \langle \hat{S}_{-\mathbf{q}A}^- | \hat{S}_{\mathbf{q}A}^+ \rangle \rangle, \\ G_4(\mathbf{q}, \omega) &\equiv \langle \langle \hat{S}_{-\mathbf{q}B}^+ | \hat{S}_{\mathbf{q}A}^+ \rangle \rangle. \end{aligned}$$

The equations of motion for GFs (8) are

$$\omega G_{s_1 s_2}^{\alpha\beta}(\mathbf{q}, \omega) = \langle [\hat{S}_{-\mathbf{q}s_1}^\alpha, \hat{S}_{\mathbf{q}s_2}^\beta] \rangle + \langle [\hat{S}_{-\mathbf{q}s_1}^\alpha, \hat{H}] | \hat{S}_{\mathbf{q}s_2}^\beta \rangle. \quad (10)$$

The commutators $[\hat{S}_{-\mathbf{q}s_1}^\pm, \hat{H}]$ are given in Appendix A.

LR [42] analyzed several approximate methods to decouple the infinite set of equations (10). Here, we adopt the two simplest ones: (1) the LSWT approximation, which is valid at $T = 0$ and is used to interpret the INS experiments, and (2) the Tyablikov self-consistent approximation that interpolates the GF in the interval between zero and the Néel temperature.

After the Tyablikov decoupling [39,42,44] of the higher-order GF

$$\langle \langle \hat{S}_{\mathbf{R}}^+ \hat{S}_{\mathbf{R}'}^z | \hat{S}_{\mathbf{q}A}^- \rangle \rangle \simeq \langle \hat{S}_{\mathbf{R}'}^z \rangle \langle \langle \hat{S}_{\mathbf{R}}^+ | \hat{S}_{\mathbf{q}A}^- \rangle \rangle, \quad (11)$$

the equations of motion (10) may be written in a matrix form

$$\begin{pmatrix} \epsilon - \epsilon_A & -m_3 & -m_1 & -m_2 \\ -m_3^* & \epsilon - \epsilon_B & -m_2^* & -m_1 \\ m_1 & m_2 & \epsilon + \epsilon_A & m_3 \\ m_2^* & m_1 & m_3^* & \epsilon + \epsilon_B \end{pmatrix} \begin{pmatrix} G_1 \\ G_2 \\ G_3 \\ G_4 \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} \quad (12)$$

or, briefly,

$$\mathbf{MG} = \mathbf{K}. \quad (13)$$

Here, $\epsilon \equiv \omega/(2m)$ and $m \equiv m_A = \langle \hat{S}_{\mathbf{R}A}^z \rangle$ is proportional to the magnetization of the sublattice A. The LSWT for $S = 1/2$ corresponds to a non-self-consistent theory with $m = S = 1/2$. At low temperatures, $T \ll T_N$, for the external field smaller than the spin-flop field, $B < B_c$, the magnetization of a monodomain sample is zero. Consequently, the magnetization of the sublattice B is opposite and has the same magnitude $m_B = \langle \hat{S}_{\mathbf{R}B}^z \rangle = -m_A$. Then, the other elements of the matrix $\mathbf{M}$ are

$$\epsilon_A \equiv m_0 + \mathbf{b}, \quad \epsilon_B \equiv m_0 - \mathbf{b}, \quad (14)$$

$$\mathbf{b} \equiv g_z \mu_B B / (2m), \quad (15)$$

$$m_0 \equiv \frac{1}{2} \left[\sum_{\mathbf{g}} (I_{\mathbf{g}} e^{i\mathbf{q}\mathbf{g}} - I_{\mathbf{g}}^z) + \sum_{\mathbf{f}} I_{\mathbf{f}}^z \right], \quad (16)$$

$$m_1 \equiv \frac{1}{4} \sum_{\mathbf{g}} E_{\mathbf{g}} e^{i\mathbf{q}\mathbf{g}}, \quad (17)$$

$$m_2 \equiv \frac{1}{2} \sum_{\mathbf{f}} I_{\mathbf{f}} e^{i\mathbf{q}\mathbf{f}}, \quad (18)$$

$$m_3 \equiv \frac{1}{4} \sum_{\mathbf{f}} E_{\mathbf{f}} e^{i\mathbf{q}\mathbf{f}}. \quad (19)$$

Solving the system (12) by Cramer's rule, we obtain

$$G_1(\mathbf{q}, \epsilon) = \frac{Z(\epsilon)}{\det \mathbf{M}}, \quad (20)$$

where $Z(\epsilon) \equiv \det \mathbf{M}'$, with

$$\mathbf{M}' \equiv \begin{pmatrix} \epsilon - \epsilon_B & -m_2^* & -m_1 \\ m_2 & \epsilon + \epsilon_A & m_3 \\ m_1 & m_3^* & \epsilon + \epsilon_B \end{pmatrix}. \quad (21)$$

In the model of Fig. 1, the matrix elements are expressed in terms of the following parameters:

$$m_0 = I_4[\cos(\pi L) - 1] + 2(I_1^z + I_2 + I_3), \quad (22)$$

$$m_1 = 0, \quad (23)$$

$$m_2 = 2 \cos(\pi H) \cos(\pi K) e^{4i\pi z_{\text{Cu}} L} (I_1 - I_2 e^{-i\pi L} + I_3 e^{-2i\pi L}), \quad (24)$$

$$m_3 = \cos(\pi H) \cos(\pi K) e^{4i\pi z_{\text{Cu}} L} (I_1^x - I_1^y). \quad (25)$$

The Cartesian components of the quasimomentum $\mathbf{q}$ are given in the reciprocal-lattice units of the *structural* unit cell, i.e., $\mathbf{q}\mathbf{a} \equiv 2\pi H$, $\mathbf{q}\mathbf{b} \equiv 2\pi K$, and $\mathbf{q}\mathbf{c} \equiv 2\pi L$. We recall that in Bi_2CuO_4 the structural unit cell is twice as large as the magnetic one.

The determinants in Eq. (20) are

$$\begin{aligned} \det \mathbf{M} &= \epsilon^4 - \epsilon^2(\epsilon_A^2 + \epsilon_B^2 - 2|m_2|^2 + 2|m_3|^2) \\ &\quad + \epsilon_A^2 \epsilon_B^2 - 2\epsilon_A \epsilon_B (|m_2|^2 + |m_3|^2) + |m_2^2 - m_3^2|^2 \end{aligned} \quad (26)$$

$$= (\epsilon^2 - \epsilon_+^2)(\epsilon^2 - \epsilon_-^2), \quad (27)$$

$$\det \mathbf{M}' = (\epsilon^2 - \epsilon_A^2 + |m_2|^2)(\epsilon + \epsilon_A) - |m_3|^2(\epsilon - \epsilon_A). \quad (28)$$

The roots $\pm\epsilon_{\pm}$ of the equation $\det \mathbf{M} = 0$ determine the positions of the poles of the Green's function. Positive roots give the energies of spin excitations, $\omega_{\pm} = 2m\epsilon_{\pm}$. The residues at the poles $Z(\pm\epsilon_{\pm})$ are important for the calculation of the Néel temperature.

III. EXCITATION SPECTRUM IN THE LINEAR SPIN-WAVE APPROXIMATION

The linear spin-wave approximation is valid at low temperatures, $T \ll T_N$ [45–51]; see also Chap. IV of Ref. [39] and Appendix B. It corresponds to $m = S = 1/2$, so the excitation energies coincide with $\epsilon_{\pm}$, the positive roots of the biquadratic equation (26):

$$\epsilon_{\pm} = \sqrt{E_m^2 \pm 2R^2}, \quad (29)$$

$$\begin{aligned} E_m^2 &\equiv \frac{\epsilon_A^2 + \epsilon_B^2}{2} - |m_2|^2 + |m_3|^2 \\ &= b^2 + E_{\text{ea}}^2 + |m_3|^2, \end{aligned} \quad (30)$$

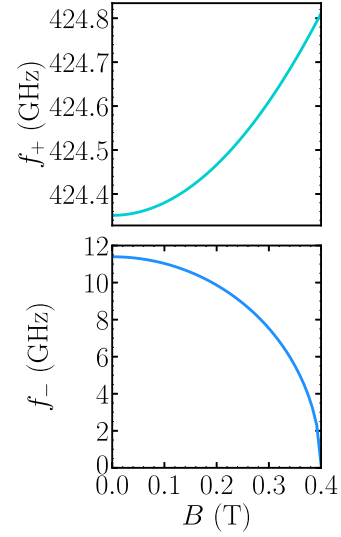


FIG. 2. AFMR frequency dependence on an external field smaller than the spin-flop field, $B < B_c$.

$$R^2 = \sqrt{(E_{\text{ea}} b)^2 + (m_0 |m_3|)^2 + (m_2 m_3^* - m_2^* m_3)^2 / 4}, \quad (31)$$

$$E_{\text{ea}}^2 \equiv m_0^2 - |m_2|^2. \quad (32)$$

The subscript “ea” abbreviates “easy axis” because E_{ea} is the excitation dispersion for the easy-axis XXZ model, where $I^z > I^y = I^x$ or, in other words, $\alpha_{\parallel} = \alpha_{\perp}$. Then, $m_3 = 0$, and the energy dependence on the external field becomes linear, so Eq. (29) acquires the form [43,49]

$$\epsilon_{\pm, \text{ea}} = E_{\text{ea}} \pm b = E_{\text{ea}} \pm g_z \mu_B B. \quad (33)$$

In the easy-axis model, the two branches are degenerate for $B = 0$.

Equation (29) shows that in the XYZ model, for $\alpha_{\parallel} \neq \alpha_{\perp}$, the spin-wave energies depend nonlinearly on the external field B . The gaps at $\mathbf{q} = 0$, i.e., at the Brillouin zone (BZ) center Γ , determine the two AFMR frequencies for an external field smaller than the spin-flop field, $B < B_c$ (see Fig. 2):

$$\Delta_{\pm} = 2\pi \hbar f_{\pm}, \quad (34)$$

$$\Delta_{\pm}^2 = b^2 + E_{\Gamma, \text{ea}}^2 + m_{3\Gamma}^2 \pm 2\sqrt{(E_{\Gamma, \text{ea}} b)^2 + m_{0\Gamma}^2 m_{3\Gamma}^2}, \quad (35)$$

$$E_{\Gamma, \text{ea}}^2 \equiv m_{0\Gamma}^2 - |m_{2\Gamma}|^2, \quad (36)$$

where the extra subscript “ Γ ” indicates that $\mathbf{q} = 0$, i.e.,

$$m_{0\Gamma} \equiv 2(I_1^z + I_2 + I_3), \quad (37)$$

$$m_{2\Gamma} \equiv 2(I_1 + I_2 + I_3), \quad (38)$$

$$m_{3\Gamma} \equiv (I_1^x - I_1^y). \quad (39)$$

Dispersion near the BZ center at zero field

In the close vicinity of the BZ center, $|H|$, $|K|$, and $|L| \ll 1/2$, we expand the expressions (22)–(25) and substitute the approximate formulas (A7)–(A9) derived in Appendix A 2

into Eq. (29). Then, the dispersion at Γ reads

$$\begin{aligned}\epsilon_{\pm} &\approx \sqrt{\Delta_{\pm}^2 + v_{H,\pm}^2 Q_{\parallel}^2 + v_{L,\pm}^2 L^2}, \\ Q_{\parallel} &= \sqrt{H^2 + K^2},\end{aligned}\quad (40)$$

where the explicit expressions for the coefficients $v_{H,\pm}$, $v_{L,\pm}$ are given in Appendix A 2 [see Eqs. (A11) and (A12)].

In the general case, when all the exchange parameters are different, $I_i^z > I_i^y \neq I_i^x$, both branches of the XYZ model have gaps at zero field. In the present model, they are

$$\begin{aligned}\Delta_+ &= 2\sqrt{[I_1^y + I_1^z + 2(I_2 + I_3)](I_1^z - I_1^x)} \\ &= 2\sqrt{2I(\alpha_{\perp} + \alpha_{\parallel})(I + I_2 + I_3)},\end{aligned}\quad (41)$$

$$\begin{aligned}\Delta_- &= 2\sqrt{[I_1^x + I_1^z + 2(I_2 + I_3)](I_1^z - I_1^y)} \\ &= 2\sqrt{2I\alpha_{\parallel}[I(2 - \alpha_{\perp} + \alpha_{\parallel}) + 2(I_2 + I_3)]},\end{aligned}\quad (42)$$

where we have taken into account that $I_1^z > I_1^y > I_1^x > 0$, i.e., $|I_1^x - I_1^y| = (I_1^y - I_1^x)$.

In the isotropic limit $I_1^z = I_1^y = I_1^x$ (XXX-model, $\alpha_{\parallel} = \alpha_{\perp} = 0$), both branches become gapless. They have acousticlike dispersions along the high-symmetry directions near Γ :

$$\epsilon_+^{\text{iso}}(H, K, 0) = \epsilon_-^{\text{iso}}(H, K, 0) \approx v_H^{\text{iso}} Q_{\parallel}, \quad (43)$$

$$\epsilon_+^{\text{iso}}(0, 0, L) = \epsilon_-^{\text{iso}}(0, 0, L) \approx v_L^{\text{iso}} L. \quad (44)$$

In the easy-plane case $I_1^z = I_1^y$ (XXZ model, $\alpha_{\parallel} = 0$, $\alpha_{\perp} > 0$), one of the branches is gapless and has an acousticlike dispersion near Γ ,

$$\epsilon_-^{\text{ep}}(H, K, 0) \approx v_{H,-}^{\text{ep}} Q_{\parallel}, \quad (45)$$

$$\epsilon_-^{\text{ep}}(0, 0, L) \approx v_{L,-}^{\text{ep}} L. \quad (46)$$

In contrast, in the easy-axis case $I_1^x = I_1^y$ (XXZ model, $\alpha_{\parallel} = \alpha_{\perp} > 0$), the two branches coincide and have a gap

$$\Delta_+^{\text{ea}} = \Delta_-^{\text{ea}} = 4\sqrt{I\alpha_{\parallel}(I + I_2 + I_3)}. \quad (47)$$

IV. ANISOTROPY PARAMETERS

The anisotropy parameters may be found from the knowledge of the two AFMR frequencies at an external field smaller than the spin-flop field, $B < B_c$. In Ref. [33], the zero-field resonance frequency was reported to be $f_+ \approx 390$ GHz. It corresponds to a large gap of $\Delta_+ \approx 1.6$ meV, which is close to the value $\Delta_+ \approx 1.75$ meV measured in the INS experiment [14]. However, the determination of the second, low resonance frequency $f_- \sim 10$ GHz is hindered by a large linewidth [30–32]. Because of this, we will use another way to find the small in-plane anisotropy parameter $\alpha_{\parallel}$.

When the external field is applied along the direction of the ordered moment, one of the gaps [Eq.(35)] shrinks and the other one widens (see Fig. 3). The narrow gap $\Delta_-(b)$ vanishes at a critical value

$$\begin{aligned}b_c &= g_z \mu_B B_c = \sqrt{m_{0\Gamma}^2 - (|m_{2\Gamma}| + |m_{3\Gamma}|)^2} \\ &= 4\sqrt{\alpha_{\parallel} I(I + I_2 + I_3)}.\end{aligned}\quad (48)$$

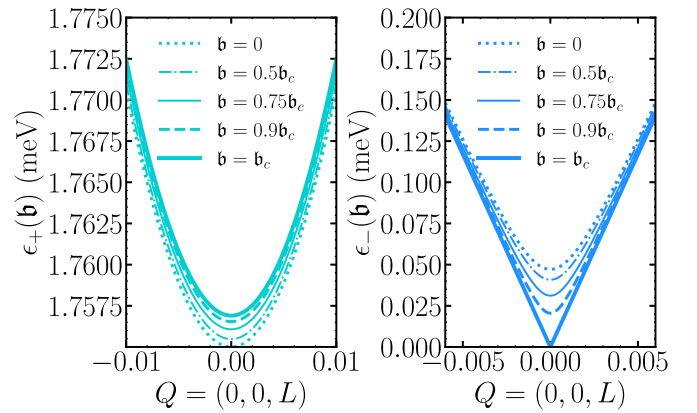


FIG. 3. Excitation dispersions near the Brillouin zone center at different external field $b = g_z \mu_B B$. Left: The branch ϵ_+ of the spectrum goes up with the increase of the field. Right: The branch ϵ_- goes down. The gap in ϵ_- vanishes at the critical value of the spin-flop transition given by Eq. (48). The model parameters are indicated in Fig. 1 caption.

Thus, substituting the value $B_c \approx 0.4$ T [14,21,35] into Eq. (48), we find the in-plane anisotropy

$$\alpha_{\parallel} = \frac{(g_z \mu_B B_c)^2}{16I(I + I_2 + I_3)} \approx 4.7 \times 10^{-6}. \quad (49)$$

Using the value of Δ_+ obtained from Eq. (41), we find the out-of-plane anisotropy parameter

$$\alpha_{\perp} \approx \frac{\Delta_+^2}{8I(I + I_2 + I_3)} - \alpha_{\parallel} \approx 0.013, \quad (50)$$

which is very close the value found in Ref. [14].

The higher AFMR frequency is $f_+ = \Delta_+/h \approx 424$ GHz, which is in good agreement with the experimental value 390 GHz [33]. The lower one, $f_- = \Delta_-/h \approx 11$ GHz, is in accord with the estimate of Ref. [30] (see Fig. 12 of that paper).

V. CALCULATION OF NÉEL TEMPERATURE

Linear spin-wave approximation is not applicable at the temperatures comparable with the ordering temperature T_N . The Tyablikov self-consistent approximation [Eq. (11)] interpolates the GF in the interval between zero and the Néel temperature. Expression (20) for the GF can be recast as a sum of four poles:

$$\begin{aligned}G_1(\mathbf{q}, \epsilon) &= \frac{Z(\epsilon)}{(\epsilon^2 - \epsilon_+^2)(\epsilon^2 - \epsilon_-^2)} \\ &= \frac{Z(\epsilon)}{2R^2} \left[\frac{1}{\epsilon_+} \left(\frac{1}{\epsilon - \epsilon_+} - \frac{1}{\epsilon + \epsilon_+} \right) \right. \\ &\quad \left. - \frac{1}{\epsilon_-} \left(\frac{1}{\epsilon - \epsilon_-} - \frac{1}{\epsilon + \epsilon_-} \right) \right].\end{aligned}\quad (51)$$

The equation for the sublattice magnetization is given by

$$\frac{1}{2} - m = \frac{1}{N} \sum_{\mathbf{q}} \langle \hat{S}_{\mathbf{q}\mathbf{A}}^- \hat{S}_{-\mathbf{q}\mathbf{A}}^+ \rangle, \quad (52)$$

where the correlation function $\langle \hat{S}_{\mathbf{qA}}^- \hat{S}_{-\mathbf{qA}}^+ \rangle$ is related to the GF [38–41]

$$\langle \hat{S}_{\mathbf{qA}}^- \hat{S}_{-\mathbf{qA}}^+ \rangle = \int_{-\infty}^{+\infty} \frac{d\omega}{e^{\beta\omega} - 1} \left(-\frac{1}{\pi} \text{Im} G_1(\mathbf{q}, \omega + i0) \right). \quad (53)$$

Equations (52) and (53) close the RPA self-consistency loop. From Eq. (51), we obtain

$$\begin{aligned} & -\frac{1}{\pi} \text{Im} G_1(\mathbf{q}, \omega + i0) \\ &= \frac{mZ(\omega/2m)}{2R^2} \left\{ \frac{1}{\epsilon_+} [\delta(\omega - 2m\epsilon_+) - \delta(\omega + 2m\epsilon_+)] \right. \\ & \quad \left. - \frac{1}{\epsilon_-} [\delta(\omega - 2m\epsilon_-) - \delta(\omega + 2m\epsilon_-)] \right\}. \quad (54) \end{aligned}$$

Then, Eq. (52) becomes

$$\begin{aligned} \frac{1}{2} - m &= \frac{1}{N} \sum_{\mathbf{q}} \frac{m}{R^2} \left\{ \frac{1}{\epsilon_+} \left[\frac{Z(\epsilon_+)}{e^{2\beta m\epsilon_+} - 1} - \frac{Z(-\epsilon_+)}{e^{-2\beta m\epsilon_+} - 1} \right] \right. \\ & \quad \left. - \frac{1}{\epsilon_-} \left[\frac{Z(\epsilon_-)}{e^{2\beta m\epsilon_-} - 1} - \frac{Z(-\epsilon_-)}{e^{-2\beta m\epsilon_-} - 1} \right] \right\}. \quad (55) \end{aligned}$$

At the Néel temperature, the sublattice magnetization vanishes, $m \rightarrow 0$. For small m , Eq. (55) reduces to

$$\frac{1}{2} - m = \frac{1}{N} \sum_{\mathbf{q}} \left[\frac{Z(\epsilon_+) + Z(-\epsilon_+)}{2\beta R^2 \epsilon_+^2} - \frac{Z(\epsilon_-) + Z(-\epsilon_-)}{2\beta R^2 \epsilon_-^2} \right] \quad (56)$$

and provides the formula for T_N :

$$k_B T_N = 1/F, \quad (57)$$

$$\begin{aligned} F &= \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{R^2} \left[\frac{Z(\epsilon_+) + Z(-\epsilon_+)}{\epsilon_+^2} - \frac{Z(\epsilon_-) + Z(-\epsilon_-)}{\epsilon_-^2} \right] \\ &= \frac{1}{N} \sum_{\mathbf{q}} \left[\frac{\epsilon_A}{\epsilon_+^2} \left(1 + \frac{|\mathbf{m}_3|^2}{R^2} \right) + \frac{\epsilon_A}{\epsilon_-^2} \left(1 - \frac{|\mathbf{m}_3|^2}{R^2} \right) \right]. \quad (58) \end{aligned}$$

As usual, in the thermodynamic limit $N \rightarrow \infty$, we pass from summation to integration over the magnetic BZ. Approximate expressions (40) may be used for analytic integration near the BZ center, where small values of ϵ_- are encountered in the denominator of the integrand. The calculation gives $T_N \approx 52$ K, which is slightly larger than the experimental value $T_N^{\text{exp}} \sim 42\text{--}47$ K.

VI. DISCUSSION AND CONCLUSION

Figure 4 shows that within the experimental accuracy of ~ 0.06 meV the dispersion $\epsilon_{\pm}(\mathbf{q})$ [Eq. (29)] coincides with that found in the INS experiments and fitted by $\omega_{\pm}(\mathbf{q})$ given by Eq. (4) of Ref. [14]. A tiny deviation from $\omega_{\pm}$ may be seen in the vicinity of Γ because of the gap in the acousticlike branch ϵ_- (see the inset in Fig. 4). The gap $\Delta_- \approx 0.047$ meV is caused by the in-plane anisotropy characterized by the parameter $\alpha_{\parallel} \approx 4.7 \times 10^{-6}$. In Appendix B, we demonstrate that within the LSWT the GF approach is equivalent to the traditional one, which is based on the approximation of the replacement of the spin-deviation operators by the

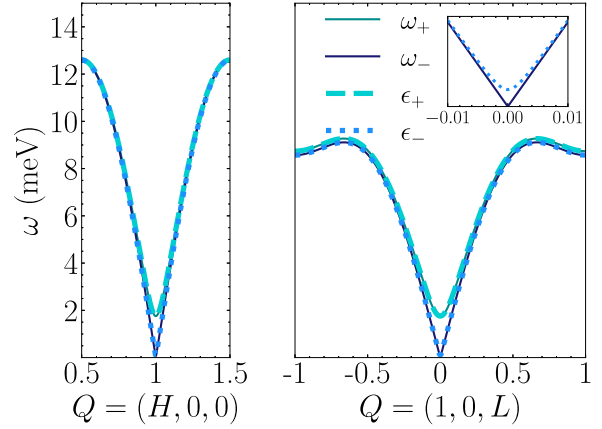


FIG. 4. Comparison of spin-wave dispersion $\epsilon_{\pm}$ [Eq. (29)] for zero field, $b = 0$ (dashed and dotted lines), with the dispersion $\omega_{\pm}$ given by Eq. (4) of Ref. [14] (solid lines). The inset shows the vicinity of the Brillouin zone center Γ , where the acousticlike branch ϵ_- has a gap caused by the in-plane anisotropy [see Eq. (40)]. The parameter set is the same as in Fig. 3.

bosonic ones; see Refs. [45–51] and Chap. IV of Ref. [39] and references therein. Both methods yield the same spectrum $\epsilon_{\pm}(\mathbf{q})$ [Eq. (29)].

The authors of Ref. [14] argue that the tetragonal symmetry of Bi_2CuO_4 demands that the acousticlike branch of the dispersion be gapless. However, this statement contradicts their observation that the anisotropic XYZ model is allowed by symmetry in this compound. Indeed, the acousticlike branch ω_- given by Eq. (4) of Ref. [14] remains gapless at *any* value of the in-plane anisotropy, including the case $\alpha_{\parallel} = \alpha_{\perp} > 0$. However, this case corresponds to an easy-axis XXZ model $I_1^z > I_1^x = I_1^y$, which evidently has gaps at $B = 0$ in the two degenerate branches [43] [see Eq. (47)]. In Appendix C, we elucidate the origin of the discrepancy between our result for the spin-wave spectrum and that of Ref. [14].

The gap $\Delta_- = \epsilon_-(0)$ given by Eq. (42) has a strong square root dependence on $\alpha_{\parallel}$, as shown in Fig. 5.

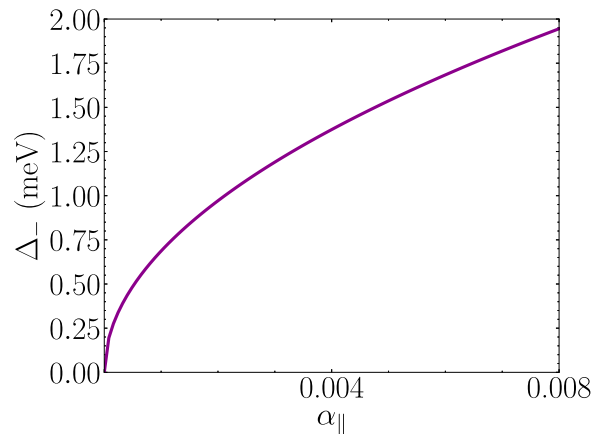


FIG. 5. The gap in the acousticlike branch $\Delta_- = \epsilon_-(0)$ given by Eq. (42) on in-plane anisotropy parameter $\alpha_{\parallel} = (I_1^z - I_1^y)/(I_1^z + I_1^y)$. Other parameters are the same as in Figs. 3 and 4.

The spin-flop transition in Bi_2CuO_4 in the field applied in the $\mathbf{ab}$ plane indicates that the gap in the acousticlike branch closes at the critical field $B_c \approx 0.4$ T (cf. Fig. 3). The estimate of the critical field in Ref. [14] is based on the relation $g\mu_B B_{c,Y} \cos \theta \sim \Lambda$. For $\Lambda = 0.014$ μeV [14] and $\cos \theta \sim 1$, it yields $B_{c,Y} \approx 1.2 \times 10^{-4}$ T $\ll B_c$. Thus, our description of the magnetism in Bi_2CuO_4 is fully consistent and is supported by the experimental data.

In Ref. [26], the Néel temperature was calculated within the Tyablikov approximation for the easy-axis XXZ model. As mentioned in the Sec. I, at that time, in 2007, there was no certainty regarding the direction of the ordered moments and the anisotropy parameters of the Bi_2CuO_4 spin Hamiltonian nor regarding the parameters of the isotropic exchange interactions. That is why previous T_N calculations were performed for three different sets of model parameters collected in rows 3, 5, and 9 of Table I in Ref. [26]: (1) Set 1 was obtained in Ref. [26] from DFT calculations; (2) Set 2 was a modification of Set 1 that fitted the INS data of Ref. [29]; and (3) Set 3 was the one originally proposed in Ref. [29]. These sets were supplemented by the anisotropy parameters that reproduced the spin gap $\Delta_{[29]} \approx 1.87$ meV reported in Ref. [29]. The results for the three models are $T_{N,1} \approx 59$ K and $T_{N,2} \approx T_{N,3} \approx 47$. The main difference between Set 2 and Set 3 consists in swapping (in our notation) $I_1 \rightleftharpoons I_3$, but it gives almost the same spectrum (cf. Fig. 8 of Ref. [26]), and hence the same Néel temperature.

The difference of our result $T_N \approx 52$ K from that of Ref. [26] is mainly due to the overall increase of the spectral width. The change of the model from the easy-axis to the easy-plane one does not strongly affect the result. As a check, we may modify our present model and set

$\alpha_{\parallel} = \alpha_{\perp} = 0.013/2$ in order to obtain an easy-axis XXZ model with two degenerate branches that coincide with ϵ_{+} . Then, the calculation according to Eq. (57) gives $T_{N,\text{ea}} \approx 54$ K. The isotropic version of our model, $\alpha_{\parallel} = \alpha_{\perp} = 0$, yields $T_{N,\text{iso}} \approx 51$ K. As expected, the anisotropy suppresses fluctuations and leads to the increase of the ordering temperature T_N when the anisotropy terms are added to the isotropic model.

We conclude that the XYZ model consistently describes the spin excitation spectrum reported in the INS experiments, the metamagnetic spin-flop transition, and the antiferromagnetic resonance. Still open remains the reinterpretation of the optical spectroscopy experiments [27–29], which were initially interpreted within the assumption that the ordered Cu spins are parallel to the crystallographic axis $\mathbf{c}$.

ACKNOWLEDGMENTS

Discussions with S.-L. Drechsler and M. D. Kuz'min are gratefully acknowledged. This work was supported by Spanish MCIU/AEI/10.13039/501100011033, Projects No. PID2022-139230NB-I00 and No. PID2022-138750NB-C22, and by the National Academy of Sciences of Ukraine (Project No. III-6-26).

DATA AVAILABILITY

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

APPENDIX A: DETAILS OF THE CALCULATIONS

1. Commutators

The commutators of operators $\hat{S}_{-\mathbf{q}A}^{\pm}$ with the Hamiltonian $\hat{H}$ are

$$[\hat{S}_{-\mathbf{q}A}^{\pm}, \hat{H}] = [\hat{S}_{-\mathbf{q}A}^{\pm}, \hat{H}_A] + [\hat{S}_{-\mathbf{q}A}^{\pm}, \hat{H}_{AB}] + [\hat{S}_{-\mathbf{q}A}^{\pm}, \hat{H}_Z], \quad (\text{A1})$$

$$[\hat{S}_{-\mathbf{q}A}^{+}, \hat{H}_A] = \frac{1}{\sqrt{N}} \sum_{\mathbf{R} \in A} e^{-i\mathbf{q}\mathbf{R}} \sum_{\mathbf{g}} \left[-I_{\mathbf{g}}^z \hat{S}_{\mathbf{R}}^{+} \hat{S}_{\mathbf{R}+\mathbf{g}}^z + I_{\mathbf{g}} \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{g}}^{+} + \frac{E_{\mathbf{g}}}{2} \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{g}}^{-} \right], \quad (\text{A2})$$

$$[\hat{S}_{-\mathbf{q}A}^{+}, \hat{H}_{AB}] = \frac{1}{\sqrt{N}} \sum_{\mathbf{R} \in A} e^{-i\mathbf{q}\mathbf{R}} \sum_{\mathbf{f}} \left[-I_{\mathbf{f}}^z \hat{S}_{\mathbf{R}}^{+} \hat{S}_{\mathbf{R}+\mathbf{f}}^z + I_{\mathbf{f}} \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{f}}^{+} + \frac{E_{\mathbf{f}}}{2} \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{f}}^{-} \right], \quad (\text{A3})$$

$$[\hat{S}_{-\mathbf{q}A}^{-}, \hat{H}_A] = \frac{1}{\sqrt{N}} \sum_{\mathbf{R} \in A} e^{-i\mathbf{q}\mathbf{R}} \sum_{\mathbf{g}} \left[I_{\mathbf{g}}^z \hat{S}_{\mathbf{R}}^{-} \hat{S}_{\mathbf{R}+\mathbf{g}}^z - I_{\mathbf{g}} \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{g}}^{-} - \frac{E_{\mathbf{g}}}{2} \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{g}}^{+} \right], \quad (\text{A4})$$

$$[\hat{S}_{-\mathbf{q}A}^{-}, \hat{H}_{AB}] = \frac{1}{\sqrt{N}} \sum_{\mathbf{R} \in A} e^{-i\mathbf{q}\mathbf{R}} \sum_{\mathbf{f}} \left[I_{\mathbf{f}}^z \hat{S}_{\mathbf{R}}^{-} \hat{S}_{\mathbf{R}+\mathbf{f}}^z - I_{\mathbf{f}} \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{f}}^{-} - \frac{E_{\mathbf{f}}}{2} \hat{S}_{\mathbf{R}}^z \hat{S}_{\mathbf{R}+\mathbf{f}}^{+} \right], \quad (\text{A5})$$

$$[\hat{S}_{-\mathbf{q}A}^{\pm}, \hat{H}_Z] = \pm g_z \mu_B H \hat{S}_{-\mathbf{q}A}^{\pm}. \quad (\text{A6})$$

Similarly, the commutators $[\hat{S}_{-\mathbf{q}B}^{\pm}, \hat{H}_B]$, $[\hat{S}_{-\mathbf{q}B}^{\pm}, \hat{H}_Z]$ are obtained from Eqs. (A2), (A4), and (A6) by the substitution $\mathbf{R} \rightarrow \mathbf{R} + \rho_B$. The commutators with $\hat{H}_{AB}$ are

$$[\hat{S}_{-\mathbf{q}B}^{+}, \hat{H}_{AB}] = \frac{1}{\sqrt{N}} \sum_{\mathbf{R} \in A} e^{-i\mathbf{q}(\mathbf{R}+\rho_B)} \sum_{\mathbf{f}} \left[-I_{\mathbf{f}}^z \hat{S}_{\mathbf{R}-\mathbf{f}+\rho_B}^{+} \hat{S}_{\mathbf{R}+\rho_B}^z + I_{\mathbf{f}} \hat{S}_{\mathbf{R}-\mathbf{f}+\rho_B}^z \hat{S}_{\mathbf{R}+\rho_B}^{+} + \frac{E_{\mathbf{f}}}{2} \hat{S}_{\mathbf{R}-\mathbf{f}+\rho_B}^z \hat{S}_{\mathbf{R}+\rho_B}^{-} \right],$$

$$[\hat{S}_{-\mathbf{q}B}^{-}, \hat{H}_{AB}] = \frac{1}{\sqrt{N}} \sum_{\mathbf{R} \in A} e^{-i\mathbf{q}(\mathbf{R}+\rho_B)} \sum_{\mathbf{f}} \left[I_{\mathbf{f}}^z \hat{S}_{\mathbf{R}-\mathbf{f}+\rho_B}^{-} \hat{S}_{\mathbf{R}+\rho_B}^z - I_{\mathbf{f}} \hat{S}_{\mathbf{R}-\mathbf{f}+\rho_B}^z \hat{S}_{\mathbf{R}+\rho_B}^{-} - \frac{E_{\mathbf{f}}}{2} \hat{S}_{\mathbf{R}-\mathbf{f}+\rho_B}^z \hat{S}_{\mathbf{R}+\rho_B}^{+} \right].$$

2. Dispersion approximation near the Brillouin zone center

Near the BZ center, $|H|$, $|K|$, $|L| \ll 1/2$, we may approximately write

$$m_0 \approx m_{0\Gamma} - I_4(\pi L)^2/2, \quad (A7)$$

$$m_2 \approx m_{2\Gamma} \left[1 - \frac{\pi^2}{2}(H^2 + K^2) - \frac{I_2 + 4I_3}{m_{20}}(\pi L)^2 \right] - 2\pi i(I_2 + 2I_3)L, \quad (A8)$$

$$m_3 \approx m_{3\Gamma} \left[1 - \frac{\pi^2}{2}(H^2 + K^2) \right]. \quad (A9)$$

Substituting these expressions into Eq. (29), we obtain for the dispersion in the center of the BZ [Eq. (40)]. The coefficients are

$$v_{H,\pm} = \pi \sqrt{m_{2\Gamma}^2 - m_{3\Gamma}^2 \mp |m_{3\Gamma}| m_{0\Gamma}} \quad (A10)$$

$$= \pi \sqrt{4(I_1 + I_2 + I_3)^2 - (I_1^x - I_1^y)^2 \pm 2(I_1^x - I_1^y)(I_1^z + I_2 + I_3)}, \quad (A11)$$

$$v_{L,\pm} = \pi \sqrt{-2I_4(I_1^z + I_2 + I_3) + 4(I_2 + 4I_3)(I_1 + I_2 + I_3) - 4(I_2 + 2I_3)^2 \pm (I_1^x - I_1^y) \left[I_4 - \frac{(I_2 + 2I_3)^2}{2(I_1^z + I_2 + I_3)} \right]}. \quad (A12)$$

APPENDIX B: LINEAR SPIN-WAVE THEORY OF Bi_2CuO_4 : QUADRATIC FORM DIAGONALIZATION

Here, we show that the traditional way to find the LSWT spectrum [45–51] (see also Chap. IV of Ref. [39]) gives the same answer as the GF approach [Eq. (29)].

LSWT starts from a classical ground state of the spin system. It is obtained by substituting spin operators in the Hamiltonian (1) by vectors of the length S ($=1/2$ for our system) and minimizing the obtained form with respect to the vectors' directions. For Bi_2CuO_4 , the minimum corresponds to a Néel state with two simple cubic sublattices A and B. The classical vectors lie in the **ab** crystallographic plane. Here, we assume that the vectors are directed along **a** + **b**. LSWT describes the dynamics of small fluctuations of the spins around their classical direction. The spin-deviation operators are represented approximately by the bosonic ones [47]:

$$\hat{S}_{\mathbf{R}+\rho_A}^+ \approx c_{\mathbf{R}_A} = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} e^{i\mathbf{q}\mathbf{R}_A} c_{\mathbf{q}}, \quad (B1)$$

$$\hat{S}_{\mathbf{R}+\rho_A}^- \approx c_{\mathbf{R}_A}^\dagger = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} e^{-i\mathbf{q}\mathbf{R}_A} c_{\mathbf{q}}^\dagger, \quad (B2)$$

$$\hat{S}_{\mathbf{R}+\rho_A}^z \approx \frac{1}{2} - c_{\mathbf{R}_A}^\dagger c_{\mathbf{R}_A}, \quad [c_{\mathbf{R}_A}, c_{\mathbf{R}'_A}^\dagger] = \delta_{\mathbf{R}_A, \mathbf{R}'_A}, \quad (B3)$$

and analogously for sublattice B:

$$\hat{S}_{\mathbf{R}+\rho_B}^+ \approx d_{\mathbf{R}_B}^\dagger = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} e^{-i\mathbf{q}\mathbf{R}_B} d_{\mathbf{q}}^\dagger, \quad (B4)$$

$$\hat{S}_{\mathbf{R}+\rho_B}^- \approx d_{\mathbf{R}_B} = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} e^{i\mathbf{q}\mathbf{R}_B} d_{\mathbf{q}}, \quad (B5)$$

$$\hat{S}_{\mathbf{R}+\rho_B}^z \approx -\left(\frac{1}{2} - d_{\mathbf{R}_B}^\dagger d_{\mathbf{R}_B}\right), \quad [d_{\mathbf{R}_B}, d_{\mathbf{R}'_B}^\dagger] = \delta_{\mathbf{R}_B, \mathbf{R}'_B}. \quad (B6)$$

Let us note that the expressions (B1)–(B6) correspond to Eq. (19) of Ref. [51] with $j = A, B$ and vectors

$$\mathbf{u}_A = \begin{pmatrix} 1 \\ i \\ 0 \end{pmatrix}, \quad \mathbf{v}_A = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}, \quad \mathbf{u}_B = \begin{pmatrix} 1 \\ -i \\ 0 \end{pmatrix}, \quad \mathbf{v}_B = \begin{pmatrix} 0 \\ 0 \\ -1 \end{pmatrix}. \quad (B7)$$

Substituting expressions (B1)–(B6) into Eqs. (5) and (6) and retaining only the terms quadratic in bosonic operators, we recast the Hamiltonian as

$$\hat{H} \approx E_{\text{cl}} + \hat{H}_{\text{SW}}, \quad E_{\text{cl}} = \frac{N}{4} \left[\sum_{\mathbf{g}} I_{\mathbf{g}}^z - \sum_{\mathbf{f}} I_{\mathbf{f}}^z \right], \quad (B8)$$

$$\begin{aligned} \hat{H}_{\text{SW}} = & \sum_{\mathbf{q}} \{ \epsilon_A c_{\mathbf{q}}^\dagger c_{\mathbf{q}} + \epsilon_B d_{\mathbf{q}}^\dagger d_{\mathbf{q}} \\ & + \frac{m_1}{2} (c_{\mathbf{q}}^\dagger c_{-\mathbf{q}}^\dagger + d_{\mathbf{q}}^\dagger d_{-\mathbf{q}}^\dagger + c_{\mathbf{q}} c_{-\mathbf{q}} + d_{\mathbf{q}} d_{-\mathbf{q}}) \\ & + m_2^* d_{\mathbf{q}}^\dagger c_{-\mathbf{q}}^\dagger + m_2 c_{-\mathbf{q}} d_{\mathbf{q}} + m_3^* d_{\mathbf{q}}^\dagger c_{\mathbf{q}} + m_3 c_{\mathbf{q}}^\dagger d_{\mathbf{q}} \}. \end{aligned} \quad (B9)$$

In this representation, the interaction terms like $c_{\mathbf{R}_A}^\dagger c_{\mathbf{R}_A} c_{\mathbf{R}'_A}^\dagger c_{\mathbf{R}'_A}$ are neglected, which is justified at low temperature when the number of excited spin waves is small, $\langle c_{\mathbf{R}_A}^\dagger c_{\mathbf{R}_A} + d_{\mathbf{R}_B}^\dagger d_{\mathbf{R}_B} \rangle \ll 1$, or, in other words, $1/2 - \langle \hat{S}_{\mathbf{R}+\rho_A}^z \rangle = 1/2 + \langle \hat{S}_{\mathbf{R}+\rho_B}^z \rangle \ll 1$.

Thus, the spin-wave Hamiltonian $\hat{H}_{\text{SW}}$ is quadratic in the Bose operators:

$$\hat{H}_{\text{SW}} = \frac{1}{2} \sum_{\mathbf{q}} (\psi^\dagger \mathbf{H}_{\mathbf{q}} \psi - \epsilon_A - \epsilon_B), \quad (B10)$$

where $\psi^\dagger \equiv (c_{\mathbf{q}}^\dagger, d_{\mathbf{q}}^\dagger, c_{-\mathbf{q}}, d_{-\mathbf{q}})$ and the matrix $\mathbf{H}_{\mathbf{q}}$ is

$$\mathbf{H}_{\mathbf{q}} = \begin{pmatrix} \epsilon_A & m_3 & m_1 & m_2 \\ m_3^* & \epsilon_B & m_2^* & m_1 \\ m_1 & m_2 & \epsilon_A & m_3 \\ m_2^* & m_1 & m_3^* & \epsilon_B \end{pmatrix}. \quad (B11)$$

Such a form is diagonalized by a Bogoliubov transformation [48,52,53], which is reduced to the diagonalization of the matrix $\mathbf{G}\mathbf{H}_{\mathbf{q}}$, where $\mathbf{G}$ is a diagonal 4×4 matrix with the first and last two entries given by 1 and -1 , respectively [14,51]. It is easy to see that $\det(\epsilon\mathbf{E} - \mathbf{G}\mathbf{H}_{\mathbf{q}}) = \det\mathbf{M}$, where $\mathbf{E}$ is the identity matrix. Thus, the eigenvalues are the roots of the equation $\det\mathbf{M} = 0$ that are given by Eq. (29).

APPENDIX C: LINEAR SPIN-WAVE THEORY OF THE SPIN-1/2 XYZ MODEL ON A SQUARE LATTICE

As we have discussed in Sec. VI, the spin-wave spectrum given by Eq. (29) differs from the spectrum by Eq. (4) of Ref. [14], as shown in the inset of Fig. 4. While the difference is small for the parameters relevant for Bi_2CuO_4 , it is physically significant. The gap in the acousticlike branch Δ_- given by Eq. (42) is proportional to the zero-field AFMR frequency f_- and is responsible for the spin-flop transition at a critical field B_c given by Eq. (48).

In Ref. [14], the spectrum has a gapless acoustic branch. As a consequence, for the explanation of the actual in-plane anisotropy, which results in the spin-flop transition, the authors propose a quantum order to disorder theory, which is in strong quantitative disagreement with the observed critical field B_c . Here, we elucidate the origin of this discrepancy.

First, we see that the matrix $\mathbf{H}_{\mathbf{q}}$ in Eq. (B11) has the same form as the matrix given by Eqs. (A11)–(A13) in Appendix 2 of Ref. [14] (recall that $\mathbf{m}_1 = 0$). It is easy to check that the spin-wave spectrum [Eq. (29)] for the isotropic model $I_1^x = I_1^y = I_1^z = I$ coincides with that given by Eq. (3) of Ref. [14]. Thus, the difference between the spectrum of the XYZ model with $I_1^z > I_1^y > I_1^x > 0$ and the one given by Eq. (4) of Ref. [14] is caused by anisotropic terms. In order to demonstrate its origin, we consider here a simplified model where all the isotropic interactions in the Hamiltonian $\hat{H}$ in Eq. (1) vanish, so that $I_2 = I_3 = I_4 = 0$. With the nearest-neighbor interaction I_1 , we are left with the model considered in Ref. [42]. We may set $z_{\text{Cu}} = 0$ because the spectrum does not depend on this parameter despite the fact that the matrix elements $\mathbf{m}_2$ and $\mathbf{m}_3$ do depend on z_{Cu} [see Eqs. (24) and (25)]. We thus arrive at an XYZ model on a square lattice (SQL) [54]. In Ref. [55], a more general model is considered, which, in addition to the magnetic anisotropy, is spatially anisotropic. Below, we compare our results with those of Sec. II of Ref. [55] by setting $v_x = v_y = w_x = w_y = (I_1^x - I_1^y)/2$.

For the SQL at $B = 0$, the matrix elements of $\mathbf{H}_{\mathbf{q}}$ are

$$\epsilon_A = \epsilon_B = \mathbf{m}_0 = 2I_1^z = 2I(1 + \alpha_{\parallel}), \quad (\text{C1})$$

$$\mathbf{m}_1 = 0, \quad (\text{C2})$$

$$\mathbf{m}_2 = (I_1^x + I_1^y)\gamma_{\mathbf{q}} = I(2 - \alpha_{\parallel} - \alpha_{\perp})\gamma_{\mathbf{q}}, \quad (\text{C3})$$

$$\mathbf{m}_3 = (I_1^x - I_1^y)\gamma_{\mathbf{q}} = I(\alpha_{\parallel} - \alpha_{\perp})\gamma_{\mathbf{q}}, \quad (\text{C4})$$

where

$$\begin{aligned} \gamma_{\mathbf{q}} &\equiv \frac{1}{4} \sum_{\mathbf{f}} e^{i\mathbf{q}\mathbf{f}} = \cos(\pi H) \cos(\pi K) \\ &= \frac{1}{2} [\cos(q_x \rho_x) + \cos(q_y \rho_y)], \quad \rho = \frac{\mathbf{a} + \mathbf{b}}{2}. \end{aligned} \quad (\text{C5})$$

Then, the model Hamiltonian $\hat{H}_{\text{SW}}$ [Eq. (B9)] coincides with Eq. (1.5) of Ref. [42] as well as with Eq. (6) of Ref. [55]. We see that the diagonal matrix element $\mathbf{m}_0$ is determined by the largest interaction I_1^z , the axis $\hat{z}$ being parallel to the ordered moment direction. The nondiagonal matrix elements are real and depend on the wave vector via the function $\gamma_{\mathbf{q}}$. So does the spectrum given by Eq. (29) for the SQL

$$\epsilon_{\pm}^{\text{SQL}} = 2\sqrt{(I_1^z \mp \gamma_{\mathbf{q}} I_1^x)(I_1^z \pm \gamma_{\mathbf{q}} I_1^y)}, \quad (\text{C6})$$

which coincides with Eq. (3.11) of Ref. [42] and with Eq. (15) of Ref. [55]. Only when $\alpha_{\parallel} = 0$, so that $I_1^z = I_1^y$, the dispersion has the form $\epsilon_{\pm}^{\text{SQL}} \propto \sqrt{1 - \gamma_{\mathbf{q}}}$ and is linear near the BZ center with the slope proportional to $Q_{\parallel}$, as in Eq. (45). In a general case, the dispersion near BZ center is given by Eq. (40), with $v_{L,\pm} = 0$ and the parameters of the lower branch

$$\Delta_-^{\text{SQL}} = 2I\sqrt{2\alpha_{\parallel}(2 - \alpha_{\perp} + \alpha_{\parallel})}, \quad (\text{C7})$$

$$v_{H,-}^{\text{SQL}} = \pi I\sqrt{2[2 - \alpha_{\perp} - \alpha_{\parallel}(3 - 3\alpha_{\perp} + \alpha_{\parallel})]}. \quad (\text{C8})$$

In the region $\Delta_-^{\text{SQL}}/v_{H,-}^{\text{SQL}} \sim 2\sqrt{\alpha_{\parallel}}/\pi \ll Q_{\parallel} \lesssim 0.25$, the dispersion is quasilinear $\epsilon_-^{\text{SQL}} \approx v_{H,-}^{\text{SQL}} Q_{\parallel}$.

The matrix elements given by Eq. (5) of Ref. [14] differ from ours given by Eqs. (C1), (C3), and (C4). The correspondence between the interaction parameters $J_{\zeta\zeta}^1$ of Ref. [14] and ours is given by the relations $J_{zz}^1 = I_1^z = I(1 - \alpha_{\perp})$, $J_{yy}^1 = I_1^y = I(1 + \alpha_{\parallel})$, and $J_{xx}^1 = I_1^x = I(1 - \alpha_{\parallel})$. For comparison, we should take the matrix elements in Eq. (5) of Ref. [14] for the case of the ordered moment along $\mathbf{a} + \mathbf{b}$, i.e., $\phi = 0$. Then, the diagonal matrix element is $C_{\mathbf{q}}^{\text{SQL}} = J_{xx}^1 + J_{yy}^1 = 2I$. Despite the statement of the authors of Ref. [14] that the z' axis is along the direction of the ordered moment, the diagonal matrix element is not proportional to the largest interaction parameter $I(1 + \alpha_{\parallel})$ and thus differs from our Eq. (C1), $C_{\mathbf{q}}^{\text{SQL}} < \mathbf{m}_0$. Also substantially different from our expressions [Eqs. (C3) and (C4)] is the wave-vector dependence of the nondiagonal matrix elements

$$\begin{aligned} E_{\mathbf{q}}^{\text{SQL}} &= Ie^{-i\pi(H+K)}(\alpha_{\parallel}\kappa_{\mathbf{q}} - \alpha_{\perp}\gamma_{\mathbf{q}}), \\ F_{\mathbf{q}}^{\text{SQL}} &= Ie^{-i\pi(H+K)}[(2 - \alpha_{\perp})\gamma_{\mathbf{q}} - \alpha_{\parallel}\kappa_{\mathbf{q}}], \\ \kappa_{\mathbf{q}} &\equiv \sin \pi H \sin \pi K. \end{aligned}$$

Thus, the dependence on the anisotropy parameters of the matrix given by Eqs. (A11)–(A13) in Appendix 2 of Ref. [14] differs from that of our matrix $\mathbf{H}_{\mathbf{q}}$ [see Eq. (B11)], which leads to a different spectrum

$$\begin{aligned} \omega_{\pm}^{\text{SQL}} &= \sqrt{(C_{\mathbf{q}}^{\text{SQL}} \pm |E_{\mathbf{q}}^{\text{SQL}}|)^2 - |F_{\mathbf{q}}^{\text{SQL}}|)^2} \\ &= I\{(2 \pm |\alpha_{\parallel}\kappa_{\mathbf{q}} - \alpha_{\perp}\gamma_{\mathbf{q}}|)^2 \\ &\quad - [(2 - \alpha_{\perp})\gamma_{\mathbf{q}} - \alpha_{\parallel}\kappa_{\mathbf{q}}]^2\}^{1/2}. \end{aligned} \quad (\text{C9})$$

Near the BZ center, the dispersion of the lower branch is linear,

$$\omega_-^{\text{SQL}} \approx vQ_{\parallel}, \quad v = \pi I\sqrt{2(2 - \alpha_{\perp})}, \quad (\text{C10})$$

and does not depend on $\alpha_{\parallel}$. However, as mentioned in the main text, for $\alpha_{\parallel} = \alpha_{\perp} > 0$ the model becomes an easy-axis XXZ model $I_1^z > I_1^x = I_1^y$ having two degenerate branches with a gap $\Delta^{\text{ca}} = 4I\sqrt{\alpha_{\parallel}}$ [see Eq. (47) and Ref. [43]].

As mentioned earlier, the spectrum of an XYZ model on an SQL given by Eq. (C6) was obtained previously in Refs. [42,55]. The present derivation is transparent and may be easily reproduced by the reader.

-
- [1] N. Plakida, *High-Temperature Cuprate Superconductors: Experiment, Theory, and Applications* (Springer, Berlin, 2010).
- [2] S.-D. Chen, M. Hashimoto, Y. He, D. Song, K.-J. Xu, J.-F. He, T. P. Devereaux, H. Eisaki, D.-H. Lu, J. Zaanen, and Z.-X. Shen, Incoherent strange metal sharply bounded by a critical doping in Bi2212, *Science* **366**, 1099 (2019).
- [3] T. Li, A short review of the recent progresses in the study of the cuprate superconductivity, *Chin. Phys. B* **30**, 100508 (2021).
- [4] V. V. Val'kov, D. M. Dzebisashvili, M. M. Korovushkin, and A. F. Barabanov, Spin-polaron concept in the theory of normal and superconducting states of cuprates, *Phys. Usp.* **64**, 641 (2021).
- [5] J. A. Sobota, Y. He, and Z.-X. Shen, Angle-resolved photoemission studies of quantum materials, *Rev. Mod. Phys.* **93**, 025006 (2021).
- [6] B. Bacq-Labreuil, B. Lacasse, A.-M. S. Tremblay, D. Sénéchal, and K. Haule, Toward an *ab initio* theory of high-temperature superconductors: A study of multilayer cuprates, *Phys. Rev. X* **15**, 021071 (2025).
- [7] J. Schlappa, K. Wohlfeld, K. J. Zhou, M. Mourigal, M. W. Haverkort, V. N. Strocov, L. Hozoi, C. Monney, S. Nishimoto, S. Singh, A. Revcolevschi, J.-S. Caux, L. Patthey, H. M. Rønnow, J. van den Brink, and T. Schmitt, Spin-orbital separation in the quasi-one-dimensional Mott insulator Sr_2CuO_3 , *Nature (London)* **485**, 82 (2012).
- [8] J. Schlappa, U. Kumar, K. J. Zhou, S. Singh, M. Mourigal, V. N. Strocov, A. Revcolevschi, L. Patthey, H. M. Rønnow, S. Johnston, and T. Schmitt, Probing multi-spinon excitations outside of the two-spinon continuum in the antiferromagnetic spin chain cuprate Sr_2CuO_3 , *Nat. Commun.* **9**, 5394 (2018).
- [9] Z. Chen, Y. Wang, S. N. Rebec, T. Jia, M. Hashimoto, D. Lu, B. Moritz, R. G. Moore, T. P. Devereaux, and Z.-X. Shen, Anomalous strong near-neighbor attraction in doped 1D cuprate chains, *Science* **373**, 1235 (2021).
- [10] S.-L. Drechsler, J. Richter, R. Kuzian, J. Málek, N. Tristan, B. Büchner, A. Moskvina, A. Gippius, A. Vasiliev, O. Volkova, A. Prokofiev, H. Rakoto, J.-M. Broto, W. Schnelle, M. Schmitt, A. Ormeci, C. Loison, and H. Rosner, Helimagnetism and weak ferromagnetism in edge-shared chain cuprates, *J. Magn. Magn. Mater.* **316**, 306 (2007).
- [11] S. Nishimoto, S.-L. Drechsler, R. Kuzian, J. Richter, and J. van den Brink, Interplay of interchain interactions and exchange anisotropy: Stability and fragility of multipolar states in spin- $\frac{1}{2}$ quasi-one-dimensional frustrated helimagnets, *Phys. Rev. B* **92**, 214415 (2015).
- [12] S. Johnston, C. Monney, V. Bisogni, K.-J. Zhou, R. Kraus, G. Behr, V. N. Strocov, J. Málek, S.-L. Drechsler, J. Geck, T. Schmitt, and J. van den Brink, Electron-lattice interactions strongly renormalize the charge-transfer energy in the spin-chain cuprate Li_2CuO_2 , *Nat. Commun.* **7**, 10563 (2016).
- [13] C. E. Agrapides, S.-L. Drechsler, and S. Nishimoto, Self-organized magnon condensation in quasi-one-dimensional edge-shared cuprates without external magnetic field, *Phys. Rev. Res.* **7**, 043051 (2025).
- [14] B. Yuan, N. P. Butch, G. Xu, B. Winn, J. P. Clancy, and Y.-J. Kim, Neutron scattering study of magnetic anisotropy in the tetragonal antiferromagnet Bi_2CuO_4 , *Phys. Rev. B* **103**, 134436 (2021).
- [15] J.-C. Boivin, D. Thomas, and J. Tréhoux, étude structurale de CuBi_2O_4 , *Bull. Minéral.* **99**, 193 (1976).
- [16] J. L. Garcia-Munoz, J. Rodriguez-Carvajal, F. Sapina, M. J. Sanchis, R. Ibanez, and D. Beltran-Porter, Crystal and magnetic structures of Bi_2CuO_4 , *J. Phys.: Condens. Matter* **2**, 2205 (1990).
- [17] K. Sreedhar, P. Ganguly, and S. Ramasesha, Investigations on Bi_2CuO_4 : A quasi-one-dimensional oxide system, *J. Phys. C* **21**, 1129 (1988).
- [18] J. P. Attfield, Exchange striction and exchange constants in Bi_2CuO_4 , *J. Phys.: Condens. Matter* **1**, 7045 (1989).
- [19] R. Troc, J. Janicki, I. Filatow, P. Fischer, and A. Murasik, Three-dimensional magnetic properties of Bi_2CuO_4 , *J. Phys.: Condens. Matter* **2**, 6989 (1990).
- [20] E. W. Ong, G. H. Kwei, R. A. Robinson, B. L. Ramakrishna, and R. B. Von Dreele, Long-range antiferromagnetic ordering in Bi_2CuO_4 , *Phys. Rev. B* **42**, 4255 (1990).
- [21] K. Yamada, K.-i. Takada, S. Hosoya, Y. Watanabe, Y. Endoh, N. Tomonaga, T. Suzuki, T. Ishigaki, T. Kamiyama, H. Asano, and F. Izumi, Three-dimensional antiferromagnetic order and anisotropic magnetic properties in Bi_2CuO_4 , *J. Phys. Soc. Jpn.* **60**, 2406 (1991).
- [22] J. Konstantinovic, G. Stanisic, M. Ain, and G. Parette, On the magnetic structure of Bi_2CuO_4 , *J. Phys.: Condens. Matter* **3**, 381 (1991).
- [23] M. Ain, G. Dhalenne, O. Guiselin, B. Hennion, and A. Revcolevschi, Magnon dispersion curve of antiferromagnetic Bi_2CuO_4 , *Phys. Rev. B* **47**, 8167 (1993).
- [24] B. Rössli, P. Fischer, A. Furrer, G. Petrakovskii, K. Sablina, V. Valkov, and B. Fedoseev, Magnetic neutron scattering in single crystal Bi_2CuO_4 , *J. Appl. Phys.* **73**, 6448 (1993).
- [25] B. Rössli, B. Fåk, M.-T. Fernández-Díaz, K. Sablina, and G. Petrakovskii, Polarization analysis of spin waves in Bi_2CuO_4 , *Physica B* **234–236**, 726 (1997).
- [26] O. Janson, R. O. Kuzian, S.-L. Drechsler, and H. Rosner, Electronic structure and magnetic properties of the spin- $\frac{1}{2}$ Heisenberg magnet Bi_2CuO_4 , *Phys. Rev. B* **76**, 115119 (2007).
- [27] M. J. Konstantinovic, Z. V. Popovic, S. D. Devic, A. Revcolevschi, and G. Dhalenne, Light scattering by magnons in Bi_2CuO_4 single crystals, *J. Phys.: Condens. Matter* **4**, 7913 (1992).
- [28] M. J. Konstantinovic and Z. V. Popovic, Effects of magnetic ordering on Raman scattering spectra of Bi_2CuO_4 , *J. Phys.: Condens. Matter* **6**, 10357 (1994).

- [29] M. J. Konstantinović, Z. Konstantinović, and Z. V. Popović, Two-magnon light scattering in Bi_2CuO_4 , *Phys. Rev. B* **54**, 68 (1996).
- [30] H. Ohta, K. Yoshida, T. Matsuya, T. Nanba, M. Motokawa, K. Yamada, Y. Endoh, and S. Hosoya, EPR and AFMR of Bi_2CuO_4 in submillimeter wave region, *J. Phys. Soc. Jpn.* **61**, 2921 (1992).
- [31] A. Pankrats, G. Petrakovskii, and K. Sablina, Microwave resonance absorption in antiferromagnet Bi_2CuO_4 , *Solid State Commun.* **91**, 121 (1994).
- [32] A. Pankrats, D. Sobyatin, A. Vorotinov, and G. Petrakovskii, Antiferromagnetic resonance in Bi_2CuO_4 single crystals, *Solid State Commun.* **109**, 263 (1998).
- [33] H. Ohta, Y. Ikeuchi, S. Kimura, S. Okubo, H. Nojiri, M. Motokawa, S. Hosoya, K. Yamada, and Y. Endoh, High field AFMR measurement of Bi_2CuO_4 , *Physica B* **246–247**, 557 (1998).
- [34] M. Herak, M. Miljak, G. Dhalenne, and A. Revcolevschi, Easy plane anisotropy in Bi_2CuO_4 , *J. Phys.: Condens. Matter* **22**, 026006 (2010).
- [35] L. Zhao, H. Guo, W. Schmidt, K. Nemkovski, M. Mostovoy, and A. C. Komarek, Magnetically induced ferroelectricity in Bi_2CuO_4 , *Phys. Rev. B* **96**, 054424 (2017).
- [36] K. Kimura, Y. Otake, and T. Kimura, Visualizing rotation and reversal of the Néel vector through antiferromagnetic trichroism, *Nat. Commun.* **13**, 697 (2022).
- [37] T. Miyamoto, M. Tsujii, T. Kubo, N. Takamura, J. Zhang, R. Ikeda, Y. Otake, K. Kimura, T. Kimura, and H. Okamoto, Control of nonreciprocal directional dichroism in Bi_2CuO_4 using electric and magnetic fields of an intense terahertz pulse, *Phys. Rev. Lett.* **135**, 216903 (2025).
- [38] D. N. Zubarev, Double-time Green functions in statistical physics, *Sov. Phys. Usp.* **3**, 320 (1960).
- [39] S. V. Tyablikov, *Methods in the Quantum Theory of Magnetism* (Springer, New York, 1967).
- [40] D. Zubarev, V. Morozov, and G. Röpke, *Statistical Mechanics of Nonequilibrium Processes* (Akademie, Berlin, 1996), p. 375.
- [41] Y. G. Rudoy, The Bogoliubov-Tyablikov Green's function method in the quantum theory of magnetism, *Theor. Math. Phys.* **168**, 1318 (2011).
- [42] V. I. Lymar' and Y. G. Rudoi, Spectrum and correlation functions of an anisotropic Heisenberg antiferromagnet, *Theor. Math. Phys.* **21**, 990 (1974).
- [43] J. Feder and E. Pytte, Low-temperature behavior of the anisotropic Heisenberg antiferromagnet in the neighborhood of the magnetic phase boundaries, *Phys. Rev.* **168**, 640 (1968).
- [44] S. V. Tyablikov, Lagging and anticipating Green functions in the theory of ferromagnetism., *Ukr. Math. J.* **11**, 287 (1959).
- [45] F. Bloch, Zur theorie des ferromagnetismus, *Z. Phys.* **61**, 206 (1930).
- [46] J. C. Slater, Cohesion in monovalent metals, *Phys. Rev.* **35**, 509 (1930).
- [47] T. Holstein and H. Primakoff, Field dependence of the intrinsic domain magnetization of a ferromagnet, *Phys. Rev.* **58**, 1098 (1940).
- [48] N. Bogoliubov, *Lectures on Quantum Statistics* (Gordon and Breach Science Publishers, New York, London, Paris, 1967), Vol. 1.
- [49] T. Oguchi, Theory of spin-wave interactions in ferro- and antiferromagnetism, *Phys. Rev.* **117**, 117 (1960).
- [50] T. Oguchi, Critical behavior of the spin system with anisotropic exchange interactions. I., *J. Phys. Soc. Jpn.* **30**, 988 (1971).
- [51] S. Toth and B. Lake, Linear spin wave theory for single-Q incommensurate magnetic structures, *J. Phys.: Condens. Matter* **27**, 166002 (2015).
- [52] N. Bogolubov, On the theory of superfluidity, *J. Phys. (USSR)* **11**, 23 (1947), also reprinted in D. Pines, *The Many-Body Problem* (W.A. Benjamin, Inc., New York, 1961), p. 292.
- [53] J. Colpa, Diagonalization of the quadratic boson Hamiltonian, *Physica A* **93**, 327 (1978).
- [54] E. Manousakis, The spin- $\frac{1}{2}$ Heisenberg antiferromagnet on a square lattice and its application to the cuprous oxides, *Rev. Mod. Phys.* **63**, 1 (1991).
- [55] P. Rosenberg and E. Manousakis, Quantum Weyl-Heisenberg antiferromagnet, *Phys. Rev. B* **108**, 214431 (2023).