

Evidence of Magnon-Mediated Orbital Magnetism in a Quasi-2D Topological Magnon Insulator

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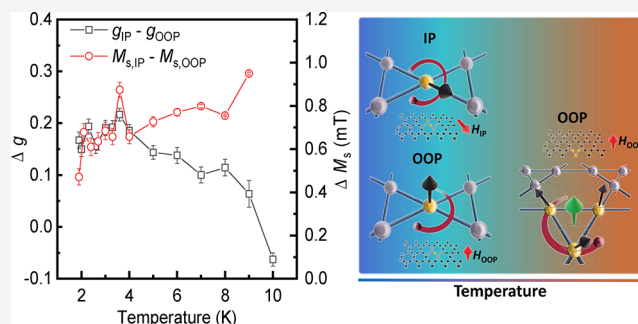
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Supporting Information

ABSTRACT: We explore spin dynamics in Cu(1,3-bdc), a quasi-2D topological magnon insulator. The results show that the thermal evolution of the Landé g factor (g) is anisotropic: $g_{\text{in-plane}}$ decreases while $g_{\text{out-of-plane}}$ increases with increasing temperature T . Moreover, the anisotropy of the g factor (Δg) and the anisotropy of saturation magnetization (ΔM_s) are correlated below 4 K, but they diverge above 4 K. We show that the electronic orbital moment contributes to the g anisotropy at lower T , while the topological orbital moment induced by thermally excited spin chirality dictates the g anisotropy at higher T . Our work suggests an interplay among topology, spin chirality, and orbital magnetism in Cu(1,3-bdc).

KEYWORDS: topological orbital moment, topological magnon insulator, kagome magnet, magnon excitation, g factor anisotropy



The discovery of topologically protected states in some systems with fermionic particles (e.g., electrons and holes) led to extensive research on topological insulators, unraveling their unique properties.¹ Such topology-protected states can also exist within the band gap of systems with bosonic quasi-particles, such as photons,^{2,3} phonons,⁴ and magnons,^{5–10} which can mediate the transport of spin and orbital angular momentum.^{11–14} While the interplay between the topology of electronic bands and spin transport properties has been intensely studied,^{15–17} the relationship between the magnonic topology and the intrinsic magnetic properties remains largely unexplored.⁷

Nontrivial magnonic band topology was predicted in magnonic crystals such as $\text{Lu}_2\text{V}_2\text{O}_7$ and $\text{Cu}(\text{bdc})^5$ (bdc = 1,3-benzenedicarboxylate). $\text{Cu}(1,3\text{-bdc})$ is a metal–organic hybrid material where Cu^{2+} ions are arranged in a geometrically perfect kagome lattice structure. The organic 1,3-bdc molecules separate the individual kagome planes, leading to weak interlayer interactions. $\text{Cu}(1,3\text{-bdc})$ is thus identified as the first quasi-two-dimensional (2D) topological magnon insulator where strong exchange coupling is confined within individual layers.¹⁸ Recent neutron scattering experiments identified flat bands originating from the unique geometry of the kagome lattice, which can be described by a Heisenberg Hamiltonian with a Dzyaloshinskii–Moriya interaction.^{19,20} Thus, these exotic properties make $\text{Cu}(1,3\text{-bdc})$ an ideal material platform for exploring the interplay between

magnonic topology and intrinsic magnetic properties, including magnetization dynamics.²¹

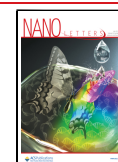
A recent theory proposed that chiral magnetism and topological magnonic excitations can be correlated with electronic orbital magnetism.^{22,23} In particular, it has been suggested that orbital magnetization can play a significant role in the dynamics of collinear antiferromagnets with weak spin–orbit coupling.²² However, experimental evidence concerning the role of the magnon-mediated electronic orbital moment in magnetization dynamics is still lacking. In this regard, it is crucial to carry out experiments to uncover the underlying physics on this topic.

Here, we use $\text{Cu}(1,3\text{-bdc})$ as a model material to study the correlation of the Landé g factor and the orbital angular momentum through vibrating sample magnetometry (VSM), a superconducting quantum interference device (SQUID), and broad-band ferromagnetic resonance (FMR) spectroscopy. The experimental results show an anisotropic gyromagnetic ratio and a corresponding g factor tensor. This anisotropy is found to be correlated with the difference in in-plane (IP) and

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out-of-plane (OOP) saturation magnetizations at lower temperatures ($T < 4$ K); it indicates a contribution from an electronic orbital moment. Surprisingly, such a correlation breaks down when $T > 4$ K. A further analysis reveals that the magnon-mediated electronic topological orbital moment has contributed to the magnetic moment at higher temperatures. We have built a foundation for elucidating the intriguing interplay of topology, spin excitations, and electronic orbital magnetism. This work also highlights the unique properties of quasi-2D topological magnon insulators for building novel spintronic devices.

It was previously shown that, in the ground state of Cu(1,3-bdc), spins within each kagome plane are ferromagnetically ordered, while spins in neighboring planes are antiferromagnetically ordered (Figure 1a).

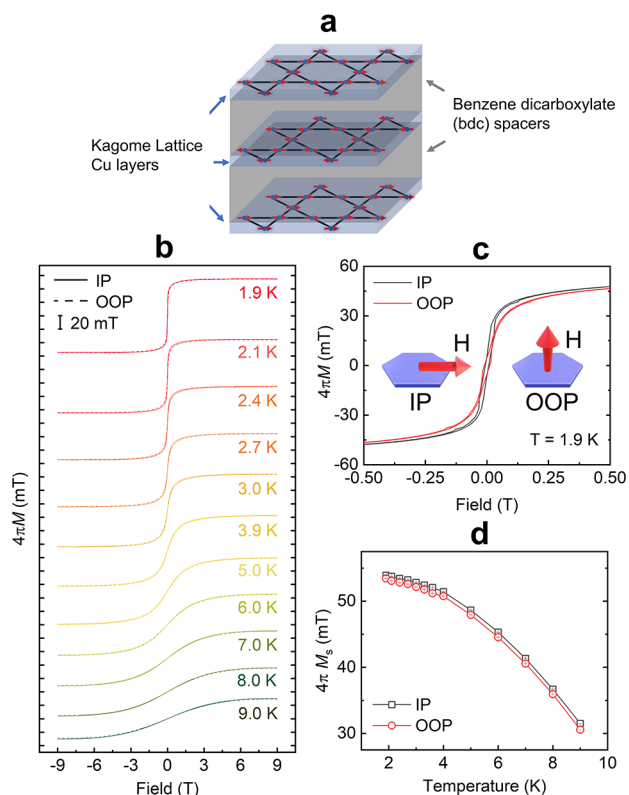


Figure 1. Quasi-static magnetic properties of Cu(1,3-bdc). (a) Schematic of Cu(1,3-bdc) structure. Kagome lattice Cu layers are spaced by benzenedicarboxylate (bdc) organic layers. Red arrows represent spins within the kagome lattice. (b) Plots of magnetization vs. externally applied field at different temperatures. The magnetic field was applied along the kagome plane (IP) of the Cu(1,3-bdc) sample (solid lines), and perpendicular (OOP) to the kagome plane (dashed lines). (c) Enlarged IP and OOP hysteresis loops at $T = 1.9$ K. The left and right insets show the IP and OOP field directions, respectively. (d) Temperature dependence of $M_{s,IP}$ (gray) and $M_{s,OOP}$ (red) extracted from the VSM measurements.

The quasi-static magnetization data of Cu(1,3-bdc) are presented in Figure 1b–d. Here, several notes should be made. First, the magnetization as a function of IP and OOP fields at temperatures ranging from 1.9 to 9 K was measured (Figure 1b). Consistent with a previous measurement,²⁰ Cu(1,3-bdc) favors a slight in-plane alignment (Figure 1c). Second, Cu(1,3-bdc) is a quasi-2D material. It is estimated that the inter-kagome-plane interaction is about 0.3% of the intra-kagome-

plane interaction. The three-dimensional Néel transition at ~ 1.8 K corresponds to the antiferromagnetic ordering of the spins on different kagome planes, which is driven by the weak interplanar interaction. The dominant intra-kagome-plane interaction, which is ferromagnetic, is responsible for the short-range ferromagnetic ordering within each plane that persists well above the transition temperature of 1.8 K.²⁰ Thus, we summarize the magnetic transitions in Cu(1,3-bdc) as follows. When $T < 1.8$ K, Cu(1,3-bdc) exhibits antiferromagnetic ordering of the spins on different kagome planes. When $1.8 \text{ K} < T < 9$ K, short-range ferromagnetic ordering exists within each plane. When $T > 9$ K, Cu(1,3-bdc) transitions into a more paramagnetic state. Third, the magnetization curves were fitted with a hyperbolic tangent function to extract the saturation magnetization M_s (refer to Note V in the Supporting Information for details). The temperature dependence of the IP and OOP saturation magnetization $M_{s,IP}$ and $M_{s,OOP}$, respectively, is plotted in Figure 1d. Intriguingly, $M_{s,IP}$ is higher than that of $M_{s,OOP}$ across all tested temperatures (1.9–9 K). For example, $M_{s,IP}$ is $\sim 0.8\%$ larger than $M_{s,OOP}$ at 1.9 K, which has been confirmed by both the VSM and SQUID measurements (refer to Note XI in the Supporting Information for details). This effect is also observed in the previous magnetization measurement.²⁰ With the hypothesis that the difference in $M_{s,IP}$ and $M_{s,OOP}$ results from the orbital magnetic moment,^{22,23} we carried out magnetization dynamics measurements to verify this understanding.

Figure 2 presents the FMR data. A schematic of the FMR measurement is shown in Figure 2a. A microwave diode and lock-in amplifier were used for signal detection. A vector network analyzer (VNA) was also used to perform the FMR measurements and generate consistent data (see Note VI in the Supporting Information for details). Figure 2b,c show microwave power absorption vs. external field at microwave frequencies ranging from 3 to 27 GHz at 1.9 K along the IP and OOP directions, respectively. The FMR profiles were measured at temperatures up to 10 K. At each microwave frequency f , the FMR profiles were fitted to Lorentzian + anti-Lorentzian functions^{25,26} to extract the resonance field H_r and the full width at half-maximum (fwhm) line width ΔH_{fwhm} (see Note VIII in the Supporting Information). The resonance frequencies are plotted against the fitted IP and OOP resonance fields, as shown in Figure 2d,e. We adopt $H_r - H_{offset}$ as the horizontal axes, where $H_{offset} = 0.4$ T. These plots reveal a linear relationship between the resonance frequency and the resonance magnetic field (refer to Note VI in the Supporting Information for details). The data points can be fitted by the Kittel equations²⁶

$$f = \gamma'_{IP} \sqrt{(H_r + 4\pi M_{eff})H_r} \quad (1)$$

$$f = \gamma'_{OOP} (H_r - 4\pi H_{eff}) \quad (2)$$

Equations 1 and 2 are for the IP and OOP cases, respectively. Here, f is the microwave frequency, $\gamma' = \frac{\gamma}{2\pi}$ is the reduced gyromagnetic ratio, and H_{eff} is the effective field representing the contribution from magnetocrystalline anisotropy. The g factor is calculated using $g = |\gamma| \frac{\hbar}{\mu_B}$, where μ_B is the Bohr magneton and $\hbar$ is the reduced Planck's constant.

Figure 2f plots the temperature dependence of the g factor (left Y axis) and γ' (right Y axis) along the IP and OOP fields, respectively. Several observations can be made from Figure 2f.

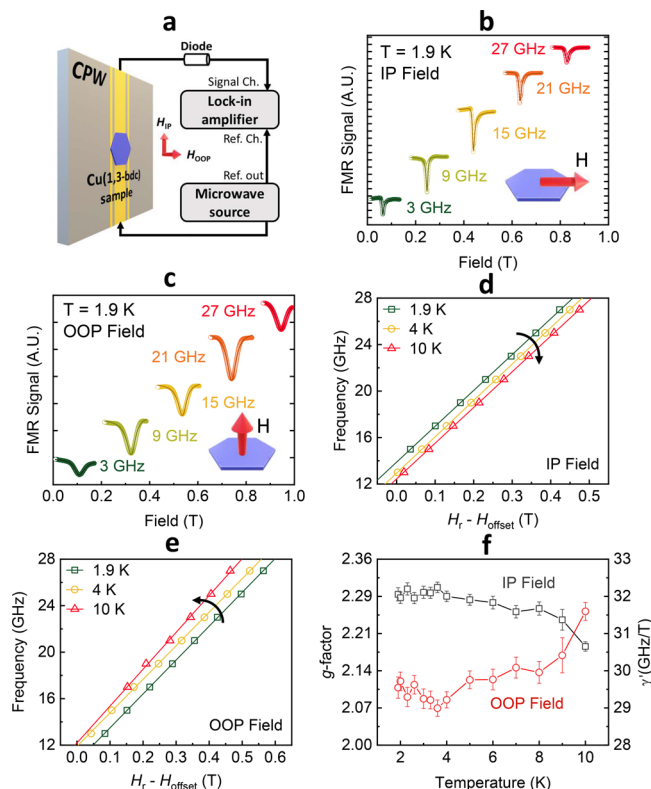


Figure 2. Broad-band FMR spectroscopy and analysis. (a) Schematic of the experimental setup. (b, c) FMR profiles measured at different microwave frequencies at $T = 1.9$ K for IP and OOP fields, respectively. Both the data points and fitted curves are presented. The insets in (b) and (c) show the IP and OOP field directions, respectively. (d, e) Resonance frequency f vs resonance field $H_r - H_{\text{offset}}$ at 1.9 K, 4 K, and 10 K for external fields applied IP and OOP, respectively. Here, $H_{\text{offset}} = 0.4$ T. The data points are fitted to the Kittel equations (1) and (2), respectively. The arrows denote the behavior of increasing temperature. (f) Temperature dependence of the Landé g factor (left axis) and gyromagnetic ratio γ (right axis) for external fields along the IP (gray) and OOP (red) directions.

First, in both the IP and OOP cases, the g factor deviates from $g = 2$, which is for a free electron without orbital momentum. This deviation indicates an orbital contribution to the magnetic moment in Cu(1,3-bdc).^{27,28} Second, g_{IP} is significantly larger than g_{OOP} at lower temperatures (i.e., < 9 K). Third, the thermal evolution of g_{IP} and g_{OOP} shows opposite trends: g_{IP} decreases with increasing temperature, while g_{OOP} increases with increasing temperature.

Both the quasi-static and dynamic magnetization measurements suggest the footprint of the electronic orbital moment. Thus, the difference of the g factor $\Delta g = g_{\text{IP}} - g_{\text{OOP}}$ is plotted together with the difference of the saturation magnetization $\Delta M_s = M_{s,\text{IP}} - M_{s,\text{OOP}}$ in Figure 3a. Figure 3a shows two contrasting behaviors: a low-temperature regime where the behavior of Δg is similar to that of ΔM_s with no significant deviation at $T < 4$ K and a high-temperature regime where Δg deviates greatly from ΔM_s at $T > 4$ K.

It is thus of interest to interpret the temperature-dependent anisotropy of the g factor in terms of the dynamics of orbital magnetism. At low temperatures, the g factor anisotropy can be attributed to the anisotropy of the orbital magnetization in Cu(1,3-bdc) along the IP and OOP directions, with the IP case having a greater orbital contribution. The deviation of Δg from

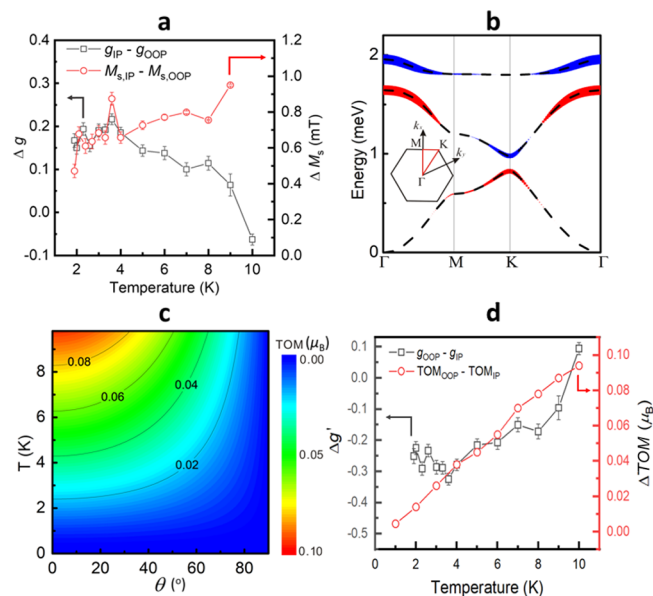


Figure 3. Electronic topological orbital moment in Cu(1,3-bdc). (a) Temperature dependence of Δg (plotted on the left Y axis) and ΔM_s (plotted on the right Y axis). (b) Flat band analysis for the magnonic bands of Cu(1,3-bdc). Red and blue represent the positive and negative signs of the local topological orbital moment (TOM) L^{TOM} , respectively. The line thickness denotes the corresponding magnitude. The insert represents the first Brillouin zone. The red lines connect the high-symmetry points that are selected in the dispersion. (c) TOM magnitude vs temperature and polar angle θ . $\theta = 0^\circ$ (90°) means that magnetization is along the OOP (IP) direction. The colors represent the magnitude of the integrated TOM in μ_B . The magnetic field B is assumed to be zero. (d) $\Delta g'$ (left Y axis) and ΔTOM (right Y axis) as a function of temperature.

ΔM_s at higher temperatures comes as a surprise, because they are expected to be closely correlated at all temperatures. Thus, this points to a different mechanism for the orbital correction of the g factor; it cannot be explained by the orbital motion of electrons around atomic cores.

A recent theoretical work uncovered contributions to the electronic orbital magnetism originating from spin chirality generated from spin disorder.^{13,29} This new development in the area of thermally driven spin fluctuations points out a route to explain the observed behavior in Cu(1,3-bdc). According to the theory, the nonvanishing net spin chirality can arise even in a collinear fluctuating spin system in its ground state. This can be directly translated into a topological electronic orbital motion, the strength of which is given by the topological orbital susceptibility relating the degree of the chirality to the magnitude of the orbital magnetization.³⁰ The magnitude of the topological orbital susceptibility can be sizable in materials with weak spin–orbit coupling, i.e. Cu(1,3-bdc), as shown in conventional microscopic calculations. Furthermore, in ferromagnetic kagome systems with nontrivial topological magnonic bands, the spin chirality mediated by thermal fluctuations can imprint sizable electronic orbital magnetization; their sign and magnitude are controlled by the parameters of the studied system.¹³

Following the approach in ref 13, we use the extracted parameters which describe the spin-exchange interactions in Cu(1,3-bdc)¹⁹ to compute the magnonic properties. The calculation yielded thermally induced net chirality and

corresponding orbital magnetization in the system. Specifically, we use the effective spin Hamiltonian of Cu(1,3-bdc) in eq 3:

$$H = -\frac{1}{2} \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{2} \sum_{ij} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) - B \cdot \kappa^{\text{TO}} \sum_{ijk} \hat{\mathbf{e}}_{ijk} [\hat{\mathbf{S}}_i \cdot (\hat{\mathbf{S}}_j \times \hat{\mathbf{S}}_k)] - \mu_B g_e B \cdot \sum_i \mathbf{S}_i, \quad (3)$$

where J_{ij} mediates the Heisenberg exchange between spins $\mathbf{S}_i$ and $\mathbf{S}_j$ on sites i and j in the first term. The second term is the antisymmetric Dzyaloshinskii–Moriya interaction (DMI) quantified by vectors $\mathbf{D}_{ij}$. The fourth term couples the spins to an external magnetic field $\mathbf{B}$. The third term is the ring-exchange interaction term, which explicitly describes the Zeeman interaction of the topological orbital moment (TOM, $\mathbf{L}^{\text{TOM}}$) with the external magnetic field $\mathbf{B}$.¹³ TOM marks a special type of electronic orbital moment. This term is given by the product of the scalar spin chirality (SSC) and the topological orbital susceptibility κ^{TO} .^{29–32} Due to the symmetry of the planar kagome lattice, the TOM and DMI vectors are both perpendicular to the kagome plane (OOP), in the same direction as the applied external magnetic field $\mathbf{B}$. In eq 3, the spin operator $\mathbf{S}_i$ at site i is set as $\frac{1}{2}$, g_e is set as 2, the nearest-neighbor exchange interaction J is set as 0.6 meV and the DMI vector is set as $D_{ij} = (0, 0, 0.09)$ meV. The topological orbital susceptibility κ^{TO} is set as a typical value of $-0.2 \mu_B$, as suggested by previous studies.^{29–32}

In Figure 3b, we plot the magnonic-band-resolved contributions to the orbital moment in Cu(1,3-bdc) for $B = 0$. One can see a strong correlation between the orbital moment and the topological band inversion of magnonic bands at low energies and between the first and second modes. Then, we integrate the contribution of the band-resolved orbital moment to the overall orbital moment of Cu(1,3-bdc) at finite temperatures. Figure 3c presents the calculated TOM as a function of temperature and polar angle θ that indicates the magnetization direction. From Figure 3c, two observations are made. First, the analysis shows that the symmetry of the system allows orbital magnetization originating from spin excitations in the OOP case but prohibits it in the IP case. Second, the value of the thermally induced orbital moment increases monotonously with temperature and becomes sizable when $T > 4$ K along the OOP direction. In Figure 3d, the differences of the g factor ($\Delta g' = g_{\text{OOP}} - g_{\text{IP}}$) and ($\Delta \text{TOM} = \text{TOM}_{\text{OOP}} - \text{TOM}_{\text{IP}}$) are plotted as a function of temperature. One can see that the $\Delta g'$ and ΔTOM values deviate below 4 K but become strongly correlated above 4 K. Thus, our theory accurately predicts the general trend and the change in the sign of $\Delta g'$ observed experimentally, as shown in Figure 3d. These facts suggest that the spin excitations can renormalize the fundamental quantum mechanical constant conventionally associated with the atomic orbital magnetism. To this end, our findings have provided experimental evidence of a new mechanism for orbital dynamics in topological magnonic systems.

Figure 4 summarizes all the orbital contributions to the magnetic moment. At low temperatures, the orbital contributions come from the motions of electrons orbiting their atomic cores for both IP and OOP cases. The black arrows in the left two illustrations represent the summation of the spin and the orbital angular momentum of the orbiting electron (red) around the nucleus (yellow). When temperature increases, the

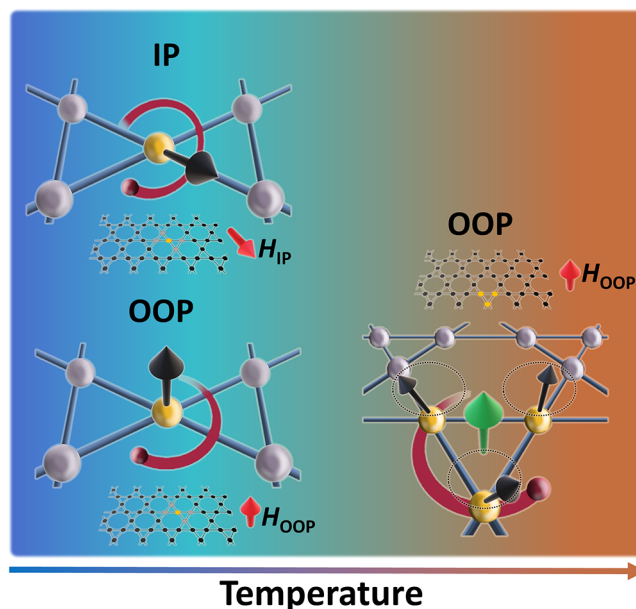


Figure 4. Orbital magnetic moment. The left two illustrations show the electronic orbital moment occurring at low temperatures. The black arrows represent the overall spin and orbital moment for a field applied IP (top left) and OOP (bottom left). The right illustration shows the magnon-mediated topological orbital moment (green arrow) arising from thermally driven scalar spin chirality.

thermal fluctuations give rise to spin chirality allowed by the symmetry of the kagome lattice in the OOP case (as shown in the right illustration). In this scenario, an electron (red) hops between a precessing noncollinear spin triplet (yellow), generating TOM (green arrow). As the temperature increases, magnon scattering, such as magnon–magnon scattering and magnon–phonon scattering, is supposed to cause the IP and OOP orbital moment to decay. This is why the g factor along the IP direction decreases at higher temperatures, as shown in Figure 2f. In this regard, the increase of the g factor in the OOP direction with increasing temperature highlights the contribution of TOM in the Cu(1,3-bdc) system.

It is helpful to identify the limiting case of the TOM effect: namely, the saturation temperature of TOM. The linear spin-wave theory that we have used cannot predict the saturation temperature of TOM. However, we have explored the g factor above 10 K using the FMR technique. Our results show that the FMR signals become significantly weaker and noisier above 10 K. We have estimated the g_{OOP} and g_{IP} parameters, which decay and approach $g = 2$ beyond 12 K. This indicates that TOM may saturate around 12 K.

In summary, anisotropies of the Landé g factor and saturation magnetization are observed in Cu(1,3-bdc). When $T < 4$ K, the differences of g factor (Δg) and saturation magnetization (ΔM_s) are correlated, which indicates the contribution of the orbital moment of electrons to the magnetic moment. The deviation of Δg and ΔM_s at $T > 4$ K can be explained by the fact that spin chirality mediated by thermal fluctuations induces sizable electronic orbital magnetization. Our work has highlighted Cu(1,3-bdc) as an important material platform to understand the interplay of topology, spin excitations, and orbital magnetism, thereby presenting a potential direction for establishing new material platforms for building novel spintronic devices. Moreover, our study has pointed out a new way to probe the orbital moment in

quantum magnets via g factor measurements. Future work that studies the chiral and topological orbital magnetism of domain walls and skyrmions³³ as well as skyrmion–topological magnon interactions in Cu(1,3-bdc) is of great interest.⁷

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.2c00562>.

Details of material preparation and characterization, ferromagnetic resonance measurements, SQUID measurements, data fittings, and theoretical calculations (PDF)

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Notes

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