Field-induced quantum spin disordered state in spin-1/2 honeycomb magnet Na$_2$Co$_2$TeO$_6$

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Spin-orbit coupled honeycomb magnets with the Kitaev interaction have received a lot of attention due to their potential of hosting exotic quantum states including quantum spin liquids. Thus far, the most studied Kitaev systems are 4$d$/5$d$-based honeycomb magnets. Recent theoretical studies predicted that 3$d$-based honeycomb magnets, including Na$_2$Co$_2$TeO$_6$ (NCTO), could also be a potential Kitaev system. Here, we have used a combination of heat capacity, magnetization, electron spin resonance measurements alongside inelastic neutron scattering (INS) to study NCTO’s quantum magnetism, and we have found a field-induced spin disordered state in an applied magnetic field range of $7.5 \text{T} < B (\perp \text{b-axis}) < 10.5 \text{T}$. The INS spectra were also simulated to tentatively extract the exchange interactions. As a 3$d$-magnet with a field-induced disordered state on an effective spin-1/2 honeycomb lattice, NCTO expands the Kitaev model to 3$d$ compounds, promoting further interests on the spin-orbital effect in quantum magnets.
Magnetic systems due to their relatively strong SOC. The examples include H₂Li₂Ir₂O₆, α-Li₂IrO₃, α-Na₂IrO₃, and α-RuCl₃. H₂Li₂Ir₂O₆ was initially thought to be a QSL, but later studies argue for a random singlet state resulted from the quenched disorder. For a random singlet state induced by the mobile protons. As for the mobile protons, the magnetic ordering at the Debye temperature tends to reduce the residual entropy rather than enhance it.

Another noteworthy feature is the field dependence of the magnetization at 17 K (the insert of Fig. 1c), reaches a maximum near 9 T. This non-monotonic behavior of magnetization with increasing fields mirrors that of the magnetic specific heat Cₘ/ₜ/T (Fig. 1a), which suggests the energy gap of the magnetic excitations closes at first and then reopens with increasing fields. It is worth mentioning that this field-dependent non-monotonic behavior was also observed in the α-RuCl₃ as considered a signal of entering the field-induced non-Abelian QSL state.

Figure 2 shows the DC magnetic susceptibility χ(T) measured by the zero-field cooling process on a single crystalline sample with B ⊥ b-axis in the ab-plane. In low fields, three anomalies at T₀, Tₘ, and Tₙ are observed as shown in Fig. 2a, which mirrors the specific heat anomalies. With increasing fields, the peak with the antiferromagnetic (AFM) characteristics at T₀ shifts to lower temperatures and becomes flattened, and the other two anomalies become weaker. Thus, the temperature-independent heat at 0 T measured on the polycrystalline sample shows a sharp peak around 6 T, whereas it increases with a temperature above 10 T. The dM/dB curves at different temperatures are all well approximated by a power law at 10 T. We also note that for temperatures above 6 K and 7.5 T < B < 10.5 T, when the system is in the disordered state as suggested by the specific heat and susceptibility data, the dM/dB curves qualitatively coincide with each other.

We interpret our magnetization data heuristically as follows. Between 7.5 and 10.5 T, the system is in a spin-disordered state with short-range spin correlations. On the one hand, the short-range spin correlations increase the dM/dB intensity with increasing temperatures since these thermal fluctuations thermally activate the spins that can be flipped by the magnetic field. On the other hand, the increasing temperature thermalizes the already polarized spins to decrease the dM/dB intensity. Therefore, the temperature-dependent dM/dB intensity below 6 K and 7.5 T < B < 10.5 T indicates there exist strong short-range spin correlations for such low temperatures in the spin-disordered state. Above 6 K and 7.5 T < B < 10.5 T, the thermal fluctuations quickly thermalize the already polarized spins so that the dM/dB intensity decreases with increasing temperature. By contrast, the system crosses over to the polarized state above 10.5 T. Thus, the temperature-independent dM/dB intensity at 10.5 T reflects a characteristic field at which these two competing effects compensate each other. We, therefore, take 10.5 T as the crossover field from the correlated spin-disordered state to the spin-polarized state. The magnetization curve further suggests that the saturation field for the α-RuCl₃ b-axis case is around Bₛ = 12.5 T with saturation magnetization Mₛ = 2.01μ₀/C₀ at 2.5 K obtained by subtracting off the Van-Vleck magnetic contribution.

High-field electron spin resonance (ESR). As shown in Fig. 3, the high-field ESR data measured at 2 K exhibits a rich excitation
spectrum, in which the modes A–C were observed in the ordered phase and the mode D was only detected with \( B > 6 \) T. The modes A–C can be described by conventional AFM resonance modes and single-magnon excitations at the \( \Gamma \) point\(^{33,30} \), the related intensities become weaker and disappear above \( T_N \) with the increasing temperature (Supplementary Fig. 3). The inset of Fig. 3k shows the electron paramagnetic resonance (EPR) spectra measured at 50 K with 214 GHz. From the resonance fields obtained by Supplementary S(2), the calculated g-factors are \( g_{ab} = 4.13 \) and \( g_c = 2.3 \), respectively. These values are also consistent with the magnetization data. The saturation magnetization \( M_S = 2.01 \mu_B/\text{Co}^{2+} \) for \( B \perp b \)-axis, Fig. 2c, leads to a \( g_B = 4.02 \) for the pseudospin-1/2 case. Both the ESR and the magnetization data corroborate the effective spin-1/2 model for \( \text{Co}^{2+} \) ions. The higher energy crystal field levels of \( \text{Co}^{2+} \) are thermally inactive in the temperature range considered in this work. The strongly anisotropic g-factors for \( \text{Co}^{2+} \) ions in the octahedral environment confirm the strong SOC and magnetocrystalline anisotropy, which can usually drive a magnetic insulator to open a spin-wave gap. The extracted resonance data from Fig. 3a–j are summarized in the frequency-field diagram shown in Fig. 3k, and the extrapolation of the frequency-field dependences of modes A–C to zero field reveal a gap \( \Delta E \approx 100 \text{ GHz} = 0.4 \text{ meV} \).

Around \( 7–8 \) T, the AFM modes of A and B approach the EPR line with \( g_{ab} = 4.13 \), which again suggests a field-driven magnetically disordered state with paramagnetic-like behavior in the \( ab \)-plane. The field-frequency curves of the A and B modes intersects with the EPR line near 6 T, which may be heuristically interpreted as the field at which the spin gap closes. This interpretation is also consistent with the specific heat data, which suggests the gap closes near 6 T. The field-frequency curve of the C mode gradually approaches the EPR line with \( g_c = 2.3 \), which indicates that its polarization is along the c-axis.

The ESR measurement reveals another mode that is not directly connected to the aforementioned AFM resonance modes, which we dub as D mode. It only appears when \( B > 6 \) T. Its excitation energy shows a linear-field dependence with a slope of \( \approx 0.15 \text{ meV/T} \), from which we deduce an effective \( g = 2.6 \). This effective g-factor is between \( g_{ab} \) and \( g_c \). The D mode must be associated with a magnetic excitation that only exists or becomes visible in the high-field spin disordered phase. Comparing with the other three modes, the D mode is much weaker, and its linewidth is broader. Its microscopic origin is unclear at the moment; however, we note its close resemblance with the ESR data of \( \alpha-	ext{RuCl}_3 \), where new modes with linear field-energy relationship emerge in the spin disordered state\(^{26,28,33} \).

Phase diagram. A phase diagram was constructed in Fig. 4 by combining the critical temperatures and fields obtained above. There are four regimes: (i) with \( B < 6 \) T, three magnetic transitions occur with decreasing temperatures. The transition at \( T_N \) should be the one from paramagnetic to zigzag AFM ordering. The one at \( T_F \) and \( T^* \) could be the adjustments of the canting moments of the zigzag order; (ii) For \( 6 \text{ T} < B < 7.5 \text{ T} \), there is a
Fig. 2 Magnetic susceptibility of Na$_2$Co$_2$TeO$_6$ for B $\perp$ b-axis. a, b Temperature dependence of susceptibility $\chi(T)$ in NCTO measured at various magnetic fields. The inset of Fig. 2b shows the honeycomb lattices of Co viewed along the c-axis. The cartoon shows the moments to be in the basal plane and parallel to the $b$-axis$^{40, 41}$, and the TeO$_6$ octahedra sit at the center of each honeycomb unit. The first NN ferromagnetic (FM) interaction $J$ originated from a collaboration between Co–O–Co (red bond) superexchange interactions and Co–Co (blue bond) direct exchange interactions. The second NN $J_2$ has multiple superexchange interaction pathways, which may lead to a wide range of variations, mainly determined by $J$ and $J_3$. The third NN AFM interaction $J_3$ arises from the existence of the Te atom in the center of the honeycomb lattice, which leads to the unique superexchange interaction pathway Co–O–Te–O–Co (golden bond). The olive arrow refers to the direction of applied magnetic field B in the magnetization. c The isothermal magnetization $M(B)$ with the applied magnetic field B $\perp$ b, B $\parallel$ b, and B $\parallel$ c-axis at 0.5 K, respectively. The dashed line indicates the Van-Vleck paramagnetic background. d The differential isothermal magnetization as functions of fields $dM/dB$ vs. B at different temperatures with B $\perp$ b.

Fig. 3 High-field ESR of Na$_2$Co$_2$TeO$_6$. a–j Frequency-dependence of ESR at 2 K; k ESR frequency-field diagram of NCTO at 2 K. The unit conversion with meV (1 meV $\approx$ 241.8 GHz) is shown on the right axis. The inset shows the EPR spectra measured at 50 K with 214 GHz. The red lines are the fitting line by Supplementary S(2). The olive squares and magenta stars are obtained from the EPR data at 50 K.
coexistence of low-field magnetically ordered state and high-field spin disordered state; (iii) most interestingly, within 7.5 T < B < 10.5 T, the system enters a spin disordered state; (iv) with B > 10.5 T, the system begins to enter the polarized state and becomes fully saturated above 12.5 T.

Spin-wave excitations. Figure 5a presents the inelastic neutron scattering (INS) measurement with incident energy E_i = 11.4 meV for NCTO at 3 K and two bands (from 0.4 to 2.9 meV and from 5.9 to 7.1 meV) are observed. The magnetic mode shows an apparent minimum near Q = 0.72 Å⁻¹, which is close to the magnitude of the M point of the honeycomb reciprocal lattice as expected for a 2D magnetic system. Moreover, the concave shapes are observed at the scattering edges, similar to the magnon excitations observed in other honeycomb lattice magnets with zigzag AFM order of α-Na_2IrO_3 and α-RuCl_3. As shown in Fig. 5c, d, the INS spectra show a gap of 0.4 meV at the M point. This gap is comparable in energy scale with the ESR gap, which corresponds to the excitation energy at the Γ point.

The diffusion refinement suggested that the magnetic moments were along the crystallographic b-axis with a possible small canting toward the c-axis. Since the inter-layer interaction is relatively weak, NCTO could be treated as a single-layer 2D compound at a first approximation. The super–super-exchange Co–O–Te–O–Co pathway produces a significant third-neighbor exchange interaction J_3. Meanwhile, the NN Co ions can interact with each other through two 90° Co–O–Co super-exchange paths or direct AFM exchange interactions. The cancellation between the different hopping contributions from d–d and d–p–d orbits can weaken the ferromagnetic NN J_2. Together, J and J_3 can stabilize a zigzag magnetic order. The second neighbor J_2 is likely to be relatively weak in that it tends to destabilize the zigzag order. However, as J_1,2,3 is isotropic in the spin space, they cannot select the direction of the magnetic moments; this is achieved by the spin anisotropic interactions such as the K and Γ terms mentioned in the Introduction.

The linear spin-wave theory (LSWT) is performed to analyze the INS spectra with the following exchange Hamiltonians:

\[
H = \sum_{<ij>}[\mathbf{S}_i \cdot \mathbf{S}_j + KS_i^y S_j^y + \Gamma(S_i^x S_j^x + S_i^z S_j^z)] + J_3 \sum_{\langle\langle i,j\rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j
\]

where, \(<i, j>\) denotes NN sites, \(\mathbf{S}_i\) and \(\mathbf{S}_j\) are effective spin-1/2 operators at sites \(i\) and \(j\), respectively, \(\alpha\) and \(\beta\) are perpendicular to the Kitaev spin axis \(\gamma\). When the \(J_2\) and \(I\) (Ising exchange interaction) are close to zero (Supplementary 4.1 Symmetries and model), the zigzag AFM order will be more stable. Finally, the powder-averaged scattering numerical results are presented in Fig. 5b. With \(K = I = 0.125 \text{ meV}\), \(J = -2.175 \text{ meV}\), and \(J_3 = 2.5 \text{ meV}\), the calculated dispersion can reproduce qualitatively the experimental data. While the INS cannot access the excitation energy gap at the Γ point, the LSWT calculation suggests an energy gap on the same order of magnitude as the M point gap, in qualitative agreement with the ESR results.

**Discussion**

The characteristic behaviors for the field-induced spin disordered state in NCTO, such as the disappearance of the peaks observed on specific heat and \(\chi(T)\), the field dependence of magnetic entropy with a maximum near 9 T below 17 K, and the existence of low energy excitations at low temperatures indicated by the \(dM/dB\) curves, are all similar to those observed for the field-induced disordered state above about 7 T in α-RuCl_3. These behaviors have been believed to be evidence that α-RuCl_3 enters a QSL state. The ESR measurements of α-RuCl_3 also show extra modes in the field-induced disordered state with a linearly increasing energy gap with increasing magnetic fields,

**Fig. 4 Phase diagram of Na_2Co_2TeO_6.** Temperature-magnetic field phase diagram for NCTO. The stars symbol the first-ordered phase boundary. The color background is used only as a simple guide.
dynamics of NCTO while it could be challenging due to the small size of single crystals grown by the flux method. In summary, the most significant finding from our studies on NCTO presented here is a QSL-like disordered state induced under fields with $7.5 T < B < 10.5 T$. Therefore, NCTO is a novel example of an effective spin-1/2 honeycomb magnet that hosts a field-induced spin disordered state. Its origin, in addition to the related spin structure and spin dynamics, calls for future experimental work on single crystals and theoretical studies.

**Methods**

**Sample preparation and characterization.** NCTO polycrystalline was prepared by a solid-state reaction method. At first, Na$_2$CO$_3$ (Alfa, 99.997%), Co$_3$O$_4$ (Alfa, 99.9%), and TeO$_2$ (Alfa, 99.99%) were mixed in a stoichiometric molar ratio with 5% excess Na$_2$CO$_3$, and fully ground; then, the mixture was loaded in an alumina crucible and sintered at 850 °C in the air for 40 h. The high-quality single crystal was then shut down to cool to room temperature. To confirm the structure and purity of the sample, powder X-ray diffraction (XRD) measurement was performed with a HUBER imaging-plate Guinier camera 670, using Cu $K_{\alpha 1}$ radiation ($\lambda = 1.54051$ Å). The XRD patterns were refined with the Rietveld method using the conventional refinement program FullProf (Supplementary Fig. 1). The magnetic properties were checked through measurements as a function of temperature ($T$) and magnetic field ($B$) using a vibrating sample magnetometer in the physical properties measurement system (PPMS, Quantum Design). Besides, magnetization measurements were also carried out using a high-sensitive Hall sensor magnetometer$^{60-62}$ for the temperature range from 0.4 to 30 K. The heat capacity measurements were carried out using the relaxation time method in the PPMS.

**High-field ESR.** The high-field ESR measurements were performed by the high-field high-frequency electron magnetic resonance spectrometer with 25 T water-cooled resistive magnet (field sweep range: 0–25 T, frequency range: 50–690 GHz, and temperature range: 2–300 K).

**Inelastic neutron scattering.** The INS of powder NCTO was performed using the High-Resolution Chopper Spectrometer (HRC) at the J-PARC$^{63}$ The HRC delivers high-resolution and relatively high-energy neutrons for a wide range of studies on materials dynamics. Spectra were collected at various temperatures by operating in high-flux mode (energy resolution of ±2%$E_i$) with $E_i = 11.4$ meV. The INS of powder NCTO was also carried out on the recently upgraded cold-neutron direct-geometry time-of-flight spectrometer NEAT II, HZB, Deutschland$^{64,65}$. Each sample was packed in aluminum cans filled with He exchange gas. Each scan was counted for around 6 h with the incident neutron energy $E_i = 3$ Å (−9 meV) and 5 Å (−3.27 meV).

**Data availability**

The data that support the findings of this study are available from the corresponding authors upon request.

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Author contributions

G.T.L., Y.W., H.D.Z., and J.M. conceived the project. Q.H. performed the XRD measurement and analyzed the data with help from G.T.L., Q.Y.R., and G.H.W. G.T.L., J.M.S., Q.H., and G.H.W. performed magnetization and heat capacity measurements and analyzed the data with help from J.C.W., Z.X.L., L.S.W., Z.Q., J.M., and H.D.Z. Q.H., L.W., J.W.M., and H.D.Z. synthesized the samples. J.J., C.K., T.M., S.A., S.I., G.G., M.R., Z.L., and J.M. performed neutron scattering measurements and analyzed the data with help from J.G.P., G.T.L., Q.Y.R., and G.H.W. W.T., C.Y.X., L.S.L., and G.T.L. performed high-field ESR measurements. Y.W. and Y.W. performed LSWT calculations with help from X.Q.W., G.T.L., J.M., Y.W., and H.D.Z. wrote the paper with input from all other co-authors. All authors discussed the data and its interpretation.

Competing interests

The authors declare no competing interests.

Additional information

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