Neutron spectroscopy evidence for a possible magnetic-field-induced gapless quantum-spin-liquid phase in a Kitaev material α-RuCl$_3$

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As one of the most promising Kitaev quantum-spin-liquid (QSL) candidates, α-RuCl$_3$ has received a great amount of attention. However, its ground state exhibits a long-range zigzag magnetic order, which defies the QSL phase. Nevertheless, the magnetic order is fragile and can be completely suppressed by applying an external magnetic field. Here, we explore the evolution of magnetic excitations of α-RuCl$_3$ under an in-plane magnetic field, by carrying out inelastic neutron scattering measurements on high-quality single crystals. Under zero field, there exist spin-wave excitations near the M point and a continuum near the Γ point, which are believed to be associated with the zigzag magnetic order and fractional excitations of the Kitaev QSL state, respectively. By increasing the magnetic field, the spin-wave excitations gradually give way to the continuous excitations. On the verge of the critical field $\mu_0 H_c = 7.5$ T, the former vanish and only the latter is left, indicating the emergence of a pure QSL state. By further increasing the field strength, the excitations near the Γ point become more intense. By following the gap evolution of the excitations near the Γ point, we are able to establish a phase diagram composed of three interesting phases, including a gapped zigzag order phase at low fields, possibly-gapless QSL phase near $\mu_0 H_c$, and gapped partially polarized phase at high fields. These results demonstrate that an in-plane magnetic field can drive α-RuCl$_3$ into a long-sought QSL state near the critical field.

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In the past few years, α-RuCl$_3$ with the honeycomb structure has been studied extensively in the pursuit of Kitaev quantum spin liquids (QSLs), which are resulting from the bond-dependent anisotropic Kitaev interactions[1–4], different from the triangular-, kagome-, or pyrochlore-structured QSL candidates with geometrical frustration[5, 6]. Now, it is well established that the ground state of α-RuCl$_3$ is actually a zigzag ordered state[7–11]. However, taking advantage of the spatial anisotropy of the Ru^{3+} d orbitals and the close-to-ideal bond configurations, it has been shown that there exists a large Kitaev interaction between the effective spin-1/2 moments[12–17]. Due to the presence of the Kitaev interaction, the zigzag order phase is in proximity to the Kitaev QSL phase[10, 18–20], although there also exist some non-Kitaev terms that make the system deviate from the QSL phase[21–32]. This provides the opportunity that by tuning the competing interactions, the zigzag magnetic order can be suppressed and a QSL state may be achieved[33–41]. In fact, there are accumulating reports that an external magnetic field applied within the honeycomb plane can suppress the magnetic order effectively and drive the system into a magnetically disordered state, utilizing various experimental probes, including magnetization[9, 42–45], specific heat[42, 43, 46–54], neutron scattering[9, 45, 46, 55, 56], nuclear magnetic resonance[43, 49, 57, 58], thermal conductivity and thermal Hall conductivity[50, 59–66], Raman, microwave, and terahertz spectroscopy[67–72], magnetodielectric[73], magnetic torque[59], resonant torsion magnetometry[74], electron spin resonance[75, 76], and thermal expansion and magnetostriction measurements[77, 78]. Nevertheless, whether the disordered phase under field is the long-sought QSL phase[50–53, 55–68, 72, 74–79], and if it is, whether it is gapless or gapped[43, 46, 49, 55, 57], remain hotly debated. Furthermore, there are also some controversies on whether the field divides the phase diagram into two parts, or three parts with a QSL phase intermediate between the low- and high-field phases[43, 46–51, 53, 55–65, 74–79].

In this Letter, we aim to solve these problems by carrying out inelastic neutron scattering (INS) measurements on the magnetic field evolution of the magnetic excitations with finer field step of 0.5 T, higher energy resolution of 0.15 meV, and stronger field strength up to 13 T, as compared to previous INS works under fields[55, 56]. Under zero field, the magnetic excitations are composed of the spin-wave excitations associated with the zigzag magnetic order[10, 15, 80], and a continuum hypothesized to be the fractional excitations associated with the Kitaev QSL state[10, 18–20, 81], which are around the M and Γ points, respectively. Under an external magnetic field applied within the a-b plane, the spin-wave excitations around the M point are gradually suppressed and vanish around the critical field $\mu_0 H_\mathrm{c} \approx 7.5$ T, accompanying the suppression and disappearance of the zigzag magnetic order. On the other hand, the continuum near the Γ point still persists when the spin waves vanish. These results are evident that the continuum around the Γ point represents the fractional excitations associated with the QSL state, and the phase near $\mu_0 H_\mathrm{c}$ is the QSL phase. By following the gap evolution of the continuum, we can divide the phase diagram into three phases, including the low-field gapped zigzag ordered state, intermediate possibly-gapless QSL, and gapped partially polarized phase.

Single crystals of α-$RuCl_3$ were grown by the chemical vapor transport method using commercially-purchased anhydrous α-$RuCl_3$ powders[15, 81]. The plate-like crystals are shiny and black with a typical size of 60 mg for each piece. Magnetic susceptibility measurements were performed using the vibrating sample magnetometer option integrated in a Physical Property Measurement System (PPMS-9T) from Quantum Design. The results showed that the sample had a single magnetic transition temperature. The specific heat measurements were also conducted on PPMS-9T. Neutron scattering measurements were conducted on two cold-neutron triple-axis spectrometers, FLEXX located at Helmholtz-Zentrum Berlin (HZB), and SIKA located at Australian Nuclear Science and Technology Organization (ANSTO)[82], both utilizing a fixed-final-energy mode with $E_\mathrm{f} = 5.0$ meV under double-focusing conditions for both the monochromator and analyzer. The energy resolutions for both instruments were $\sim$0.15 meV (full width at half maximum). Two batches of samples, both weighed $\sim$2 g in total, were labeled as Sample I and Sample II. Sample I and II arrays consisted of 20 and 22 pieces of single crystal, respectively. The former was used for measurements on both FLEXX and SIKA, whilst the latter was only measured on SIKA. They were coaligned using a backscattering Laue x-ray diffractometer and glued onto aluminum plates by hydrogen-free Cytop grease. These crystals were well aligned so that the overall mosaic spreads were both less than 3°, as determined from the rocking scans through the $(0, 0, 3)$ and $(1, 0, 0)$ Bragg peaks. All measurements were carried out in the $(H, 0, L)$ plane with magnetic field applied along the $[\text{-}1, 2, 0]$ direction. A hexagonal structure, with the routinely-adopted lattice parameters $a = b = 5.96$ Å, and

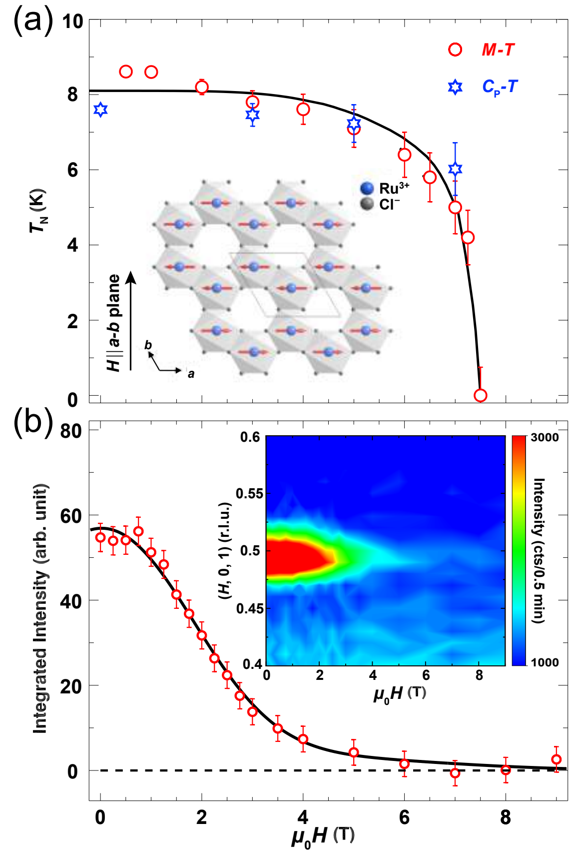

FIG. 1. (a) Dependence of the magnetic transition temperature T_N as a function of the in-plane magnetic field $\mu_0 H$ for α-$RuCl_3$, obtained from the magnetization and specific heat data. The inset shows the schematic honeycomb crystal lattice of α-$RuCl_3$ with the zigzag magnetic order. (b) Field dependence of the integrated intensities of the magnetic Bragg peak $(0.5, 0, 1)$. The inset is a contour map showing the elastic scans through $(0.5, 0, 1)$, under magnetic field applied along the $[\text{-}1, 2, 0]$ direction with strength ranging from 0 to 9 T. Black solid curves through data are guides to the eye. The errors represent one standard deviation throughout the paper.

$c = 17.20$ Å, was used throughout this Letter. The wave vector $\boldsymbol{Q}$ was expressed as (H, K, L) in reciprocal lattice unit (r.l.u.) of $(a^*, b^*, c^*) = (4\pi/\sqrt{3}a, 4\pi/\sqrt{3}b, 2\pi/c)$.

From susceptibility and specific heat measurements on α-$RuCl_3$, the relation between the magnetic transition temperature T_N and the applied in-plane magnetic field $\mu_0 H$ is obtained, as presented in Fig. 1(a). It clearly shows that T_N is decreasing with increasing field, indicating that the magnetic order is weakened and disappears at $\mu_0 H_\mathrm{c} = 7.5$ T. The magnetic field dependence of the integrated intensities for the magnetic Bragg peak $(0.5, 0, 1)$ by elastic neutron scattering measurements is plotted in Fig. 1(b). With the gradual increase of external field strength, the intensities of the Bragg peak are reduced correspondingly, also implying that the magnetic ordering is being suppressed and ultimately vanishes

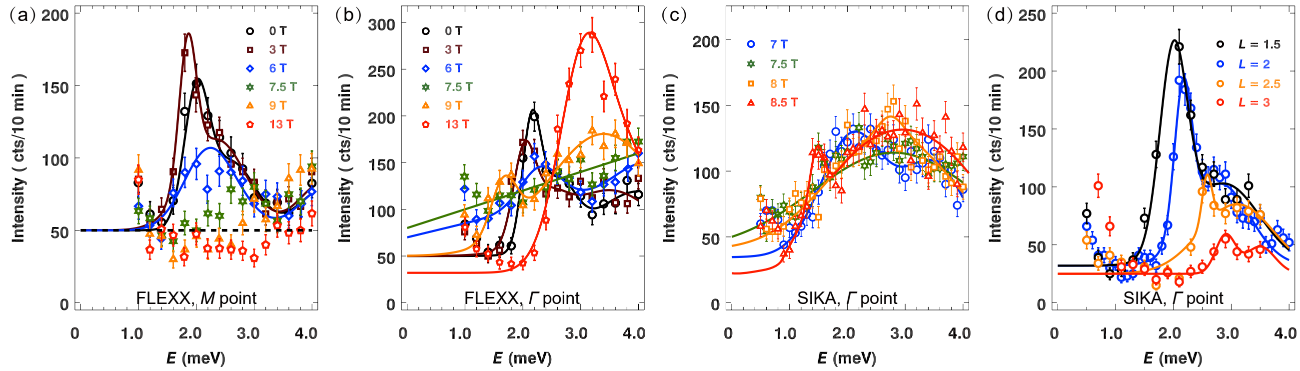

FIG. 2. (a) Constant-$\boldsymbol{Q}$ scans at the M point $(0.5, 0, 2)$ of the two-dimensional Brillouin zone, with applied field up to 13 T. (b) Same as (a) but at the Brillouin zone center Γ point $(0, 0, 2)$. (c) Similar scans as those in (b) but with finer field step of 0.5 T ranging from 7 to 8.5 T, around the critical field of 7.5 T. (d) Constant-$\boldsymbol{Q}$ scans at the Γ point $(0, 0, L)$ with different Ls under zero field. All measurements were performed on Sample I at $T = 1.8$ K. (a) and (b) were both measured on FLEXX triple-axis spectrometer while (c) and (d) were measured on SIKA. Solid lines are guides to the eye, and black dotted horizontal lines represent background signals.

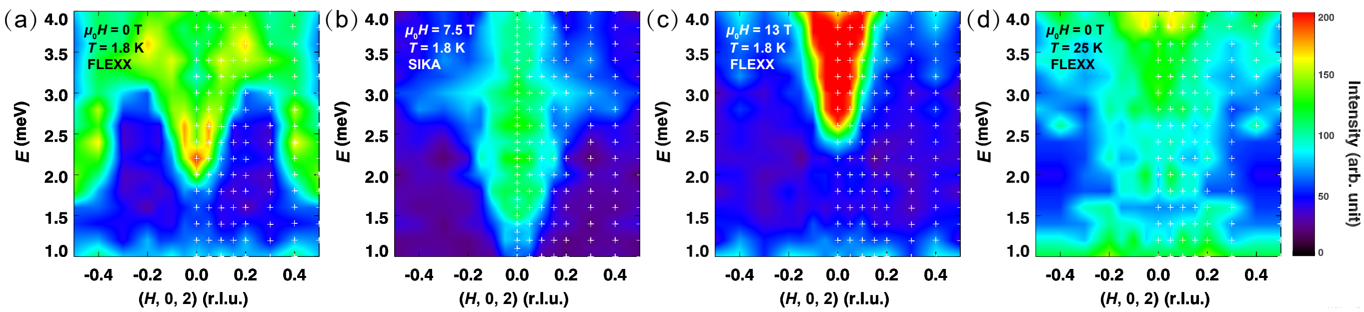

FIG. 3. Magnetic excitation spectra along the [100] direction under different magnetic fields. (a)-(c) were measured at 1.8 K, while (d) was measured at 25 K. Data of (b) were obtained from SIKA, and the rest were from FLEXX. $H = 0$ and 0.5 r.l.u. correspond to the high-symmetric Γ and M points, respectively. White cross dots mark experimental data points. In all the panels, left regions (-0.5<H<0 r.l.u.) are symmetrized from the right (0<H<0.5 r.l.u.) for better visualizing purpose. Note that according to the linear scans in Fig. 2, there are still finite intensities below 1.5 meV at the Γ point at 7.5 T, so the data in panel (b) should not be misinterpreted as there is a gap below 1.5 meV.

at around 7 T. These results are consistent with previous reports that an in-plane external field will suppress the magnetic order of α-$RuCl_3$[9, 42–45].

Based on some previous experiments[10, 15, 20, 80, 81], the magnetic excitations are basically converged at the M and Γ points in the two-dimensional Brillouin zone. Specifically, the gapped sharp excitations around the M point are the spin-wave excitations ascribed to the zigzag magnetic order, while the ones around the Γ point exhibiting broad continuous characteristics are suggested to be the fractional magnetic excitations bestowed by the proximity to the Kitaev QSL state of α-$RuCl_3$[10, 15, 18, 20, 81]. To observe the evolutions of these two types of excitations with respect to the applied field, we thus performed constant-$\boldsymbol{Q}$ scans at the M point $(0.5, 0, 2)$ and Γ point $(0, 0, 2)$. Some of the representative data are shown in Fig. 2. In Fig. 2(a), it shows that the excitations at the M point are enhanced from 0 to 3 T, which may be due to the spectral weight transfer of the magnetic Bragg peak—as shown in Fig. 1(b), the peak intensity has a great drop from 0 to 3 T. With the field further increasing, the intensity is suppressed and almost vanishes at 7.5 T and above. As shown in Fig. 2(b), the excitations at the Γ point are gradually suppressed with the field for $\mu_0 H \leq 7.5$ T, similar to those at the M point. More importantly, at 7.5 T, the peak feature of the excitations at the Γ point disappears, resulting in a featureless continuous profile in energy expected for a QSL. At 9 and 13 T, the intensities become stronger, while there are no magnetic scattering intensities at the M point for the field strength exceeding 7.5 T, indicating that the system enters a partially-polarized state, in which the magnetic moments are forced to be partially aligned with field. Furthermore, by more detailed measurements around the critical field of 7.5 T, as shown in Fig. 2(c), we can find the peak feature of the excitations at the Γ point reappears after 8 T. Overall, the featureless and continuous scan profiles at 7.5 and 8 T are quite distinct from others. At other fields, the scan reaches its background $\gtrsim$1 meV, below which the intensity raises due to the incoherent elastic scattering. On the other hand, the data points for 7.5 and 8 T are well above the background level at 1 meV, and remain finite when extended to zero energy. From these results, we can judge that the excitations are gapless at 7.5 and 8 T, but gapped at other fields, at least on a qualitative level. This issue will be discussed further in the latter part of this work.

In most previous neutron scattering experiments on α-$RuCl_3$, the L-dependence of the continuous excitations at the Γ point is typically not taken into account, and the models are normally based on the two-dimensional magnetic structure, ignoring the interlayer interactions. In Ref. 56, it has been pointed out that the excitations at the Γ point actually follow a cosine form, indicating a nonnegligible interlayer magnetic coupling. We have also measured the excitations at M and Γ points with different Ls. In Fig. 2(d), we show four representative scans at different Ls at zero field. The data show apparent L dependence, consistent with Refs. 45 and 56. Nevertheless, the evolution of the excitations with the field is similar for different Ls, as shown in the Supplementary Materials[83].

To better visualize the evolutions of the magnetic excitations with the field, we have performed a series of energy scans at various $\boldsymbol{Q}$ values, and obtained the excitation spectra along the [100] direction under different field strengths at the base temperature as plotted in Fig. 3. For comparison, the spectra at 25 K, well above the $T_{\rm N}$, are also plotted. The data in Fig. 3(b) are from SIKA, and the rest are all from FLEXX. Since the measurements were carried out on similar instruments with the same Sample I, the results are similar and comparable (See Supplementary Materials for more details[83]). Figure 3(a) clearly shows that there are two types of excitations—the spin wave excitations, with an energy gap around 1.6 meV, disperse upwards from the M point (H = $\pm$0.5 r.l.u.), and reach the band top at the Γ point; at the Γ point, there is another type of excitations. Although they seem to have a dispersion similar to the spin waves, they are shown to be incompatible with the spin waves but are a continuum representing the fractional excitations resulting from the QSL phase instead[81]. Intriguingly, as shown in Fig. 3(b), at the critical field of $\mu_0H_{\rm c} = 7.5$ T, while the spin-wave excitations at the M point completely disappear, the continuum at the Γ point persists. This strongly indicates that the disordered phase at $\mu_0H_{\rm c}$ is the long-sought QSL phase featuring fractional excitations. Furthermore, the excitations appear to extend below 1 meV and become gapless. Compared Fig. 3(b) with the spectra in the paramagnetic phase shown in Fig. 3(d), while it is similar that the spin waves both disappear, and both feature a possibly gapless continuum, the two spectra show clear differences in their boundaries of the continua, suggesting that the field-induced possibly-gapless QSL is distinct from the paramagnetic phase above $T_{\rm N}$. By further increasing the field up to 13 T, the spin waves at the M point are still absent. On the other hand, the gap at the Γ point reopens and becomes larger than that at zero field. Furthermore, the intensities of the excitations also become more intense. The gapped excitations dispersing from the Γ point look like gapped ferromagnetic spin excitations. However, the excitations of a magnetic-field-driven partially polarized state may not be necessarily consistent with those of a ferromagnetic ground state under zero field, as the excitations are dependent on the Hamiltonians. For example, in our recent work on $YbZnGaO_4$, we have found that in the fully polarized state the excitations disperse from the M point as expected for antiferromagnetic excitations, instead of from the Γ point as expected for ferromagnetic excitations, because of the presence of dominant antiferromagnetic interactions in $YbZnGaO_4$ under zero field[84]. Returning to the case of α-$RuCl_3$, we think that similar excitations of the partially-polarized and ferromagnetic states indicate the presence of a dominant ferromagnetic Kitaev interaction, as suggested in previous works[16, 17, 59, 81].

Summarizing the results in Figs. 2 and 3, and Supplementary Figs. 1 and 2, we can obtain a phase diagram of α-$RuCl_3$ based on the gap size in Fig. 4. Here, we define the energy gap as the energy where the intensity starts to rise in the low-E regime. According to the gap size, we can divide the phase diagram into three regions. The first one is the gapped zigzag ordered phase at low fields. The gap size is reduced with field, in concomitant with the suppression of the magnetic order with field as shown in Fig. 1. Around the critical field, there is a narrow regime featuring possibly gapless continuous excitations. From our measurements, we estimate the range to be about 0.5 T. By changing the way for the determination of the gap, the evolution of the gap size with the magnetic field away from the critical regime may be different, but the gapless nature at 7.5 and 8 T remains to be the same. As we show above, the magnetic order is completely suppressed here, and the excitation continuum is consistent with gapless QSL, and is therefore labeled so. With further increasing field, the magnetic moments are forced to be aligned with the field. The gap reopens and the gap size increases monotonically with field. Since the saturation field is up to above 60 T (Ref. 42), we label this phase as the gapped partially-polarized phase. We have measured the excitations with different L values. It can be seen from Fig. 4 that while the gap size has some difference at different Ls, the overall trend is similar. Such a three-zone phase diagram with an intermediate QSL phase between the low-field zigzag ordered phase and a high-field partially polarized phase featuring two quantum critical points is different from those with only two phases—a low-field zigzag ordered phase and high-field

QSL phase, divided by a quantum critical point[48, 49]. The phase space for the QSL state is also significantly narrowed down. On the other hand, it is consistent with previous literature, which features a zigzag order, QSL, and possibly topologically trivial phase[57, 61, 63, 72].

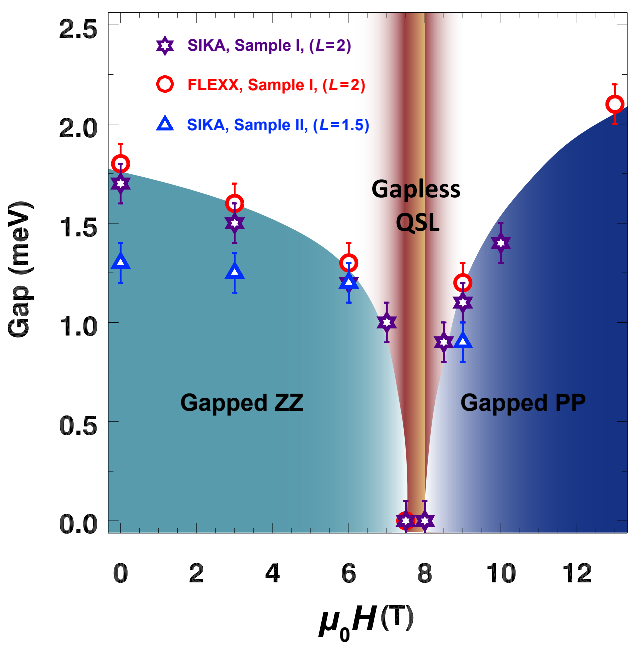

FIG. 4. Field-evolution of the energy gap and the phase diagram. The left cyan zone denotes the gapped zigzag ordered state (Gapped ZZ), central brown column represents the possibly gapless QSL state, and the right dark blue zone is the gapped partially-polarized state (Gapped PP). The purple stars and red circles denote the data measured on Sample I with $L = 2$ r.l.u. obtained from SIKA and FLEXX, respectively. The dark blue triangles represent the data measured on SIKA on Sample II with $L = 1.5$ r.l.u..

From our careful neutron scattering measurements on several batches of high-quality single crystals of α-$RuCl_3$ on different spectrometers, we have obtained comprehensive excitation spectra as well as their evolutions under external magnetic field. These results allow us to answer the three important questions we raise above. First, the disordered phase near the critical field is indeed the QSL phase. This is evident from the fact that while the spin waves associated with the magnetic order are completely suppressed, the continuum associated with the fractional excitations of the QSL state survives. Second, the QSL phase is possibly gapless. We note that in Refs. 61 and 64, a half-integer quantized plateau of the thermal Hall conductivity around the critical field was reported. This indicates the presence of a Kitaev QSL state with gapped bulk and gapless edge featuring Majorana fermions. On the other hand, another report on the thermal Hall conductivity did not observe the 1/2 plateau[63]. While this controversy remains to be solved, we believe that our direct measurements on the magnetic excitations by INS provide clear evidence that the excitations of the QSL are very likely to be gapless. Third, there is a small but finite intermediate QSL regime between the gapped zigzag order and gapped partially-polarized phase. By comparing the spectra of this field-induced gapless QSL phase [Fig. 3(b)] with those of the zero-field paramagnetic phase above T_N [Fig. 3(d)], we believe they are distinctive, although the latter phase may also feature fractional excitations[20, 85].

To summarize, we have conducted INS experiments on α-$RuCl_3$ single crystals to investigate the evolution of the magnetic excitations with an in-plane magnetic field. Our results show clearly that the spin-waves excitations around the M point are suppressed in accordance with the suppression of the magnetic order. Near the critical field of $\sim$7.5 T, the spin waves disappear but the possibly gapless continuum around the Γ point is present, indicating the emergence of a pure gapless QSL phase. Based on the evolution of the gap with field, we obtain a three-zone phase diagram, which consists of a low-field gapped zigzag order phase, an intermediate-field gapless QSL, and a high-field gapped partially-polarized state. These results constitute as evidence that an intermediate in-plane magnetic field induces a pure QSL phase in α-$RuCl_3$.

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