Signatures of low-energy fractionalized excitations in $\alpha$-RuCl$_3$
from field-dependent microwave absorption

C. Wellm, J. Zeisner, A. Alfonsov, A. U. B. Wolter, M. Roslova, A. Isava, T. Doert, M. Vojta, B. Büchner, and V. Kataev

1 Leibniz Institute for Solid State and Materials Research IFW Dresden, 01171 Dresden, Germany
2 Institut für Festkörper- und Materialphysik, Technische Universität Dresden, 01062 Dresden, Germany
3 Fachrichtung Chemie und Lebensmittelchemie, Technische Universität Dresden, 01062 Dresden, Germany
4 Institut für Theoretische Physik, Technische Universität Dresden, 01062 Dresden, Germany

Topologically ordered states of matter are generically characterized by excitations with quantum number fractionalization. A prime example is the spin liquid realized in Kitaev’s honeycomb-lattice compass model where spin-flip excitations fractionalize into Majorana fermions and Ising gauge fluxes [1]. While numerous compounds have been proposed to be proximate to such a spin-liquid phase, clear-cut evidence for fractionalized excitations is lacking. Here we employ microwave absorption measurements to study the low-energy excitations in $\alpha$-RuCl$_3$ over a wide range of frequencies, magnetic fields, and temperatures, covering in particular the vicinity of the field-driven quantum phase transition where long-range magnetic order disappears. In addition to conventional gapped magnon modes we find a highly unusual broad continuum characteristic of fractionalization which – most remarkably - extends to energies below the lowest sharp mode and to temperatures significantly higher than the ordering temperature. Our results unravel the signatures of fractionalized excitations in $\alpha$-RuCl$_3$ and pave the way to a more complete understanding of the Kitaev spin liquid and its instabilities.

Spin liquids – low-temperature states of local-moment insulators devoid of symmetry-breaking long-range order – constitute a class of most fascinating states of matter. They are characterized by topological order and fractionalization, i.e., local excitations decay into fractionalized constituents, which typically leads to continua instead of sharp modes in the dynamic response of the material. The seminal work of Kitaev has introduced a particular spin-liquid model, with compass interactions on the honeycomb lattice, whose fractionalized excitations are dispersive Majorana fermions and static Ising gauge fluxes (visons). Subsequently, it has been proposed that this model may be approximately realized in certain Mott insulators with strong spin-orbit coupling [2, 3]. Candidate materials [4] are Na$_2$IrO$_3$, different polytypes of Li$_2$IrO$_3$, and $\alpha$-RuCl$_3$. While these materials display long-range magnetic order below a small Néel temperature $T_N$, likely due to the presence of additional interactions, it is believed that they are proximate to a Kitaev spin-liquid phase. As a result, signatures of Kitaev physics are expected in various physical probes including the excitation spectrum, and pressure or magnetic field might even stabilize a spin-liquid ground state.

In $\alpha$-RuCl$_3$ [5, 6], currently considered the most promising Kitaev material, unconventional behavior has been reported in a number of experimental probes. In particular, recent inelastic neutron scattering (INS) experiments [7] have revealed an unusually broad magnetic response near the Brillouin-zone center, $q = 0$, which persists up to temperatures of 100 K, the energy scale of the estimated Kitaev coupling in $\alpha$-RuCl$_3$ [8, 9]. This elevated-energy response apparently unrelated to magnetic order may thus be consistent [7] with continuum scattering off Majorana excitations inherent in the Kitaev spin liquid [10, 11]. In addition, sharper spin-wave-like excitations at $q = 0$ with an apparent gap $\Delta_{sw} \sim 2.7$ meV at zero magnetic field have been found below $T_N = 7$ K. The existence of these magnon modes has been confirmed by other experimental techniques [12, 13] and the low-temperature study of their field dependence has revealed that $\Delta_{sw}^0(H)$ still has a sizable minimum value of $\sim 1$ meV at the critical field for the suppression of long-range AFM order $\mu_0H_c \sim 7$ T [13, 14].

To clarify the nature of the excitation spectrum of $\alpha$-RuCl$_3$ we have measured the microwave absorption (MWA) of high-quality single crystals in the temperature range $3 - 30$ K as function of the magnetic field up to 16 T, employing a high-field/high-frequency electron spin resonance (ESR) setup where the signal loss is proportional to the imaginary part of the dynamic spin susceptibility $\chi''(\omega)$ at the chosen excitation frequency $\nu = \omega/(2\pi)$ and wavector $q = 0$ (see Methods and Supplement for details). Working with fixed selected frequencies, we covered the range of $\nu = 70$ – 660 GHz (0.3 – 2.8 meV). We note that energies below 1.5 meV are practically inaccessible to INS due to the presence of the static Bragg peak and, to our knowledge, have not been addressed in detail by other studies. Our main finding is the presence of field-dependent non-resonant absorption, which is particularly strong for energies between 100 and 360 GHz (0.4 – 1.5 meV), i.e., below the apparent magnon gap, and which persists up to temperatures significantly
larger than $T_N$. This observation provides evidence for an
excitation continuum extending down to low energies of the
order $\sim 0.4\text{meV}$, likely arising from fractionalization.

\textit{Results.} Characteristic MWA spectra (signal at the
detector $S_d$) as a function of field at a given constant $\nu$
for $H \parallel \text{ab-plane}$ are summarized in Fig.1. Here, a
smaller value of $S_d$ corresponds to a larger absorption
of microwaves by the sample and vice versa. Fig.1(a)
shows the frequency dependence of the MWA signal at
3 K. At a high frequency of 480 GHz one observes a set
of lines labelled $\alpha, \beta, \gamma, \delta$ and $\epsilon$ which shift by lowering $\nu$
to 360 GHz indicating their resonant nature. These lines
are present only at $T < T_N$. Their position and the $\nu$-
dependence is in agreement with observations interpreted
in terms of resonant magnon modes at $\vec{q} = 0$ reported
in Refs. [13, 14] (Fig. S2). Remarkably, by approaching
the minimum excitation gap $\Delta_{\text{exc}} \sim 250\text{GHz}$ for these
resonance modes reported in Refs. [13, 14], the spectrum
evolves into a pronounced maximum of MWA centered at
$H_c \sim 7\text{T}$ with some residual smeared structure. This
response does not shift upon change of frequency within experi-
mental uncertainties, which suggests a non-resonant
nature of the absorption spectrum (see also Supplement,
Figs. S1 and S2).

The strong peak in the MWA spectrum in the
range $6 - 8\text{T}$ starts to develop at temperatures right below
$T_N \approx 8\text{K}$ [Fig.1(c)]. Above $T_N$, the spectra at
$\nu \leq 360\text{GHz}$ become flat for magnetic fields below $\sim 6\text{T}$
showing no field dependence of MWA [Figs.1(b) and
(c)]. Only in the vicinity of $T_N$ and at $\nu > 360\text{GHz}$ the
MWA signal still exhibits a broad bump due to strongly
broadened, rapidly decaying resonance modes $\alpha, \beta, \gamma, \delta$
and $\epsilon$ [cf. top curves in Figs.1(a) and (b)]. A remarkable
novel feature of the MWA spectrum at $T > T_N$ and
$\nu \leq 360\text{GHz}$ is the occurrence of an inflection point in
the $S_d(H)$ dependence marked by arrows in Fig.1(b)
which shifts only slightly with changing the frequency.
Above this point $S_d$ begins to increase indicating a
decrease of the MWA by the sample. Importantly, all
non-resonant MWA features are most prominent in the
frequency window $\sim 100 - 360\text{GHz}$. In particular, they
rapidly decrease at $\lesssim 150\text{GHz}$ and finally vanish below
$\sim 100\text{GHz}$ [see Fig.1(a,b) and the $T - H$ maps of the
microwave absorption plotted for different excitation
frequencies in Fig. S3 of the Supplement].
AFM denotes the antiferromagnetically ordered phase, CR marks the critical region of the enhanced spin fluctuations in the vicinity of the AFM phase, KPM stands for Kitaev paramagnet, and PG KPM denotes the (pseudo-) gapped region of a depleted density of spin excitations in the Kitaev paramagnet. Open squares in (a) are the estimates of the field-induced spin gap in temperature units from Ref. [15]. In (b) the region at higher frequencies bordered by a white-dashed line denotes the broadened resonance modes still visible at $T = 10$ K [cf. top curve in Fig. 1(b)], the grey dashed line sketches the area to which the resonance modes at $T < T_N$ are confined with the minimum spin-wave energy gap at $\mu_0H = 7$ T of $\Delta_{sw}^{\alpha} = 250$ GHz (see Fig. S2 and Refs. [13, 14]), and the horizontal white-dashed line marks the minimum energy of the excitation continuum probed by MWA. (for details see the text)

**Discussion.** Given that the microwaves probe the imaginary part of the dynamic spin susceptibility $\chi''(\omega, \vec{q} = 0)$, a strong MWA peak developing below $T_N$ around the critical field $\mu_0H_c \sim 7$ T can be related to the enhanced density of the spin fluctuations in $\alpha$-RuCl$_3$ at the vicinity of the field-induced phase transition from the AFM-ordered to a non-ordered state at higher fields. At $T > T_N$, the decrease of the MWA at higher fields ([Figs. 1(b),(c)] implies thus a depletion of the spin excitations probed by microwaves, i.e., a field-induced (pseudo-)gapped behavior of $\chi''(\omega)$ which is visible in the $S_d(H)$ spectra up to $T \sim 20 - 25$ K.

The MWA data can be used to construct a schematic phase diagram of $\alpha$-RuCl$_3$ which is drawn by lines in Fig. 2 together with the representative color-coded maps of the $S_d(H)$ spectra. Fig. 2(a) depicts the $T - \nu$ parameter space where the maps are plotted for a representative frequency $\nu = 273$ GHz (see also Fig. S3, and Fig. 2(b) presents the $\nu - H$ cut of the MWA spectra at a temperature $T = 10$ K $> T_N \approx 8$ K. We note that, with the present setup, the absorption cannot be measured on an absolute scale. Also a comparison of their values measured at different $T$ and $\nu$ is prone to a number of instrumental uncertainties. Therefore, to facilitate the pictorial presentation of the data, we plot in Fig. 2 the normalized spectra, as explained in the Methods section, so that the maps $S_d^{norm}(T, H, \nu)$ in panels (a) and (b) reflect field-induced changes in the temperature and frequency dependence of MWA.

Keeping in mind the presence of the Néel transition at $T_N \approx 8$ K at $0$ T and the sizeable spin gap $\Delta_{sw} = 40 - 50$ K [12, 17] existing at $15.5$ T, four distinct regimes enclosed by dashed lines (AFM, CR, KPM and PG KPM) with characteristic MWA properties can be identified. Given that the probing frequencies $\nu = 80 - 360$ GHz (0.3 - 1.5 meV) considered for the phase diagram in Fig. 2 are smaller than the spin-wave energies probed by INS at $q = 0$ (2.7 meV at 0 T [7] and $3.4$ meV at $8$ T [18]), the observed non-resonant effects should be related to the coupling of microwaves to spin excitations other than magnons. Such a field-dependent MWA below the spin-wave gap energy, which, as can be seen in Fig. 2(b), is most importantly present also above $T_N$ is unexpected for a conventional antiferromagnet. The non-resonant char-
acter of the MWA suggests a continuous energy spectrum of the probed excitations, typical for spin fractionalization. The vanishing of the field-dependent microwave response below $\sim 100$ GHz implies a characteristic minimum energy of $\sim 0.4$ meV (100 GHz) for this continuum [Fig. 2(b)]

At low temperatures the detected $S_H$ signal exhibits an enhanced absorption of the microwaves by the sample by approaching the critical field $\mu_0 H_c \sim 7$ T for the suppression of the AFM order. This evidences a boosted density of spin fluctuations in the critical region (CR) around this field where the spin-wave gap $\Delta_{sw} = 0$ reduces to $\sim 1$ meV [Fig. 2(b)]. In the "high"-temperature regime at low fields the interaction of microwaves with the continuum of spin excitations does not reveal a measurable field dependence. This part of the $T - H$ parameter space is denoted as correlated Kitaev paramagnet (KPM) in Fig. 2. At higher fields the MWA reduces with increasing field suggesting a depletion of the density of fractionalized excitations. This region is denoted as a pseudo-gap phase of KPM (PG KPM). Notably, the temperature range where the depletion occurs corresponds well with the energy scale of the field-induced spin gap observed in NMR [17] and thermal conductivity [15] measurements [open squares in Fig. 2(a)], whereas specific heat data reveal somewhat smaller gap values [13, 20].

In summary, our field-dependent MWA study reveals a highly unusual excitation continuum in $\alpha$-RuCl$_3$ at the wavevector $\vec{q} = 0$ at low energies between 0.4 and 1.2 meV, i.e., below the energy of the lowest reported $\vec{q} = 0$ magnon-like mode [Fig. 2(b)] and extending to temperatures $T \gg T_N$ in a broad field region up to 16 T [Fig. 2(a)]. The continuum seen by MWA gets progressively gapped above $H_c$, in line with observations of a field-induced spin gap in Refs. [15, 17, 14, 20] which value of $40 - 50$ K at 15 T is significantly larger than the energies probed by MWA. This is compliant with the MWA absorption caused by a continuum below the lowest sharp mode and persisting at elevated temperatures, a feature inconsistent with conventional magnon excitations [21]. The lower bound of the continuum appears to be quite small, of the order $\sim 100$ GHz (0.4 meV). The continuum suggests a natural explanation in terms of fractionalized excitations. Given that a generic Kitaev spin liquid displays an excitation continuum down to lowest energies [11], possibly peaked at an energy corresponding to a fraction of the Kitaev coupling [22], we consider it likely that the experimental continuum represents genuine Kitaev physics. In a fractionalization scenario, the sharper magnon-like modes observed at elevated energy may then be interpreted as bound-state magnetic excitations [14]. We emphasize that the pseudo-gapped phase above $H_c \sim 7$ T depicted in Fig. 2 as PG KPM is very unconventional in the sense that the gap seen in many physical properties [15, 17, 19, 20] is not a usual spin-wave gap typically observed in antiferromagnets or in fully polarized states of SU(2)-symmetric magnets above a saturation field, but a pseudo-gap in a continuum of excitations. Our MWA study reveals that these broad in energy excitations are of $\vec{q} = 0$ nature which offers new insights for the interpretation of other experimental results. Account for other interactions beyond the Kitaev model may bring additional complexity in the spin dynamics [23].

Methods

Crystal synthesis and characterization. Single crystals used in this study have been grown at TU Dresden. For the synthesis, pure ruthenium-metal powder (99.98 %, Alfa Aesar) was filled into a quartz ampoule under argon atmosphere, together with a sealed silica capillary containing chlorine gas (99.5 %, Riedel-de Haën). The chlorine gas was dried prior to use by passing through concentrated H$_2$SO$_4$ and CaCl$_2$, the ruthenium powder used was without further purification. The molar ratio of the starting materials was chosen to ensure in-situ formation of RuCl$_3$ and its consequent chemical transport according to the reaction: RuCl$_3$(s) + Cl$_2$ = RuCl$_4$(g) [24]. After sealing the reaction ampoule under vacuum, the chlorine-containing capillary inside was broken in order to release the gas. The ampoule was kept in the temperature gradient between 750 °C (source) and 650 °C (sink) for 5 days. The obtained crystalline product represented pure $\alpha$-RuCl$_3$ (according to powder X-ray diffraction) without inclusions of ruthenium. Single-crystal X-ray diffraction studies (Apex II diffractometer, Bruker-AXS, Mo K$\alpha$ - radiation) and EDXS (Hitachi SU 8020 SEM, 20 kV with an Oxford Silicon drift detector XMax$^N$) of the crystals have confirmed a monoclinic structure [25] and the nominal composition. The crystals are black with a shiny surface, of a size of several millimeters along the $ab$-plane, and of a thickness about 0.2 mm. MWA measurements were performed on three samples from the same batch. They all show a similar AFM ordering temperature $T_N \approx 8$ K which has been determined through magnetization measurements with a vibrating sample magnetometer (Quantum Design) with superconducting quantum interference device detection (SQUID-VSM).

Microwave absorption measurements. For measurements of MWA a home made high-frequency/high-field ESR (HF-ESR) spectrometer was employed. A vector network analyzer (PNA-X from Keysight Technologies) was used for generation and detection of microwaves in the frequency range from 75 GHz to 330 GHz. In addition, an amplifier/multiplier chain (AMC from Virginia Diodes Inc.) allowed for measurements with frequencies up to 480 GHz, detected using a hot electron InSb bolometer (QMC Instruments). The probe-head
with the sample was mounted in the variable temperature insert of the superconducting magnet system (Oxford Instruments) enabling field sweeps up to 16 T. Measurements up to 660 GHz were performed using a vector network analyser from ABmillimetre and a 14 T magnet from Cryogenic Limited. Measurements were carried out in transmission mode using a Faraday configuration, i.e. with the \( k \)-vector of the microwaves oriented parallel to the direction of the external magnetic field, which in turn was parallel to the sample \( ab \)-plane. MWA spectra at each temperature were recorded at fixed selected frequencies. (for further details see the Supplement).

The contour plots \( S_{\text{norm}}(T, H) \) presented in Fig. 2a were obtained from recorded \( S_d(H) \) spectra by first subtracting the \( S_0 \) value at a field of 15.5 T, and afterwards by subtracting the spectrum measured at 30 K from the spectra measured at other temperatures. To obtain the contour plots \( S_{\text{norm}}(\nu, H) \) shown in Fig. 2b), from all spectra except the one at \( \nu = 82 \) GHz the value \( S_d(\nu, 0 \text{T}) \) was subtracted and the spectra were normalized by a respective difference value \( \Delta S_d = [S_d(\nu, 15.5 \text{T}) - S_d(\nu, 0 \text{T})] \). To normalize the flat signal at \( \nu = 82 \) GHz the values \( S_d(\nu, 0 \text{T}) \) an \( \Delta S_d \) from the measurement at the next higher frequency \( \nu = 145 \) GHz were used.

* These authors contributed equally to this work.

[1] A. Kitaev, *Ann. Phys.* **321**, 2 (2006).
[2] G. Jackeli and G. Khaliullin, *Phys. Rev. Lett.* **102**, 017205 (2009).
[3] J. Chaloupka, G. Jackeli, and G. Khaliullin, *Phys. Rev. Lett.* **105**, 027204 (2010).
[4] S. M. Winter, A. A. Tsirlin, M. Daghofer, J. van den Brink, Y. Singh, P. Gegenwart, and R. Valenti, arXiv:1706.06113.
[5] K. W. Plumb, J. P. Clancy, L. J. Sandilands, V. Y. Shankar, Y. F. Hu, K. S. Burch, H.-Y. Kee, and Y.-J. Kim, *Phys. Rev. B* **90**, 041112 (2014).
[6] J. A. Sears, M. Songvilay, K. W. Plumb, J. P. Clancy, Y. Qiu, Y. Zhao, D. Parshall, and Y.-J. Kim, *Phys. Rev. B* **91**, 144420 (2015).
[7] A. Banerjee, J. Yan, J. Knolle, C. A. Bridges, M. B. Stone, M. D. Lumsden, D. G. Mandrus, D. A. Tennant, R. Moessner, and S. E. Nagler, *Science* **356**, 1055 (2017).
[8] L. J. Sandilands, Y. Tian, K. W. Plumb, Y.-J. Kim, and K. S. Burch, *Phys. Rev. Lett.* **114** (2015).
[9] A. Banerjee, C. A. Bridges, J.-Q. Yan, A. A. Aczel, L. Li, M. B. Stone, G. E. Granroth, M. D. Lumsden, Y. Yiu, J. Knolle, S. Bhattacharjee, D. L. Kovrizhin, R. Moessner, D. A. Tennant, D. G. Mandrus, and S. E. Nagler, *Nature Mater.* **15**, 733 (2016).
[10] J. Knolle, D. L. Kovrizhin, J. T. Chalker, and R. Moessner, *Phys. Rev. Lett.* **112**, 207203 (2014).
[11] X.-Y. Song, Y.-Z. You, and L. Balents, *Phys. Rev. Lett.* **117**, 037209 (2016).
[12] A. Little, L. Wu, P. Lampen-Kelley, A. Banerjee, S. Pantankar, D. Rees, C. A. Bridges, J.-Q. Yan, D. Mandrus, S. E. Nagler, and J. Orenstein, ArXiv e-prints (2017), arxiv:1704.07357 [cond-mat.str-el].
[13] Z. Wang, S. Reschke, D. Huvonen, S.-H. Do, K.-Y. Choi, M. Gensch, U. Nage, T. Rööm, and A. Loidl, ArXiv e-prints (2017), arxiv:1706.06157 [cond-mat.str-el].
[14] A. N. Ponomaryov, E. Schulze, J. Wosnitza, P. Lampen-Kelley, A. Banerjee, J.-Q. Yan, C. A. Bridges, D. G. Mandrus, S. E. Nagler, A. K. Kolezhuk, and S. A. Zvyagin, ArXiv e-prints (2017), arxiv:1706.07240 [cond-mat.str-el].
[15] R. Hentrich, A. U. B. Wolter, X. Zotos, W. Breig, D. Nowak, A. Isaeva, T. Doert, A. Banerjee, P. Lampen-Kelley, D. G. Mandrus, S. E. Nagler, J. Sears, Y.-J. Kim, B. Büchner, and C. Hess, ArXiv e-prints (2017), arxiv:1703.08623 [cond-mat.str-el].
[16] R. Kubo and K. Tomita, *J. Phys. Soc. Jpn.* **9**, 888 (1954).
[17] S.-H. Baek, S.-H. Do, K.-Y. Choi, Y. S. Kwon, A. U. B. Wolter, S. Nishimoto, J. van den Brink, and B. Büchner, *Phys. Rev. Lett.* **119**, 037201 (2017).
[18] A. Banerjee, P. Lampen-Kelley, J. Knolle, C. Balz, A. A. Aczel, B. Winn, Y. Liu, D. Pajerowski, J.-Q. Yan, C. A. Bridges, A. T. Savici, B. C. Chakoumakos, M. D. Lumsden, D. A. Tennant, R. Moessner, D. G. Mandrus, and S. E. Nagler, ArXiv e-prints (2017), arxiv:1706.07003 [cond-mat.mtrl-sci].
[19] J. A. Sears, Y. Zhao, Z. Xu, J. W. Lynn, and Y.-J. Kim, *Phys. Rev. B* **95**, 180411 (2017).
[20] A. U. B. Wolter, L. T. Corredor, L. Janssen, K. Nenkov, S. Schönecker, S.-H. Do, K.-Y. Choi, R. Albrecht, J. Hunger, T. Doert, M. Vojta, and B. Büchner, *Phys. Rev. B* **96**, 041405 (2017).

[21] We note that the MWA continuum is unlikely to be explained by a coupling of one-magnon and two-magnon states in the ordered phase. First, the field-dependent MWA is observed also at temperatures significantly larger than \( T_N \) and, second, at energies below the spin-wave gap at wavevector \( q = 0 \) (i.e. at the \( \Gamma \)-point of the Brillouin zone). Second-order effects which might yield sub-gap spin excitations at the \( \Gamma \)-point, e.g., due to the softening of the spin waves at the AFM-ordering wavevector at the \( \Gamma \)-point, appear unlikely as they would result in small-intensity features only. This conclusion is consistent with recent theory work [20].

[22] J. Knolle, G.-W. Chern, D. Kovrizhin, R. Moessner, and N. Perkins, *Phys. Rev. Lett.* **113**, 187201 (2014).
[23] S. M. Winter, K. Riedel, D. Kaib, R. Coldea, and R. Valenti, ArXiv e-prints (2017), arxiv:1707.08144 [cond-mat.str-el].
[24] M. Binnewies, R. Claum, M. S. Schmidt, and P. Schmidt, *Chemical Vapor Transport Reactions* (Walter de Gruyter GmbH & Co. KG, Berlin.Boston, 2012).
[25] R. D. Johnson, S. C. Williams, A. A. Haghighirad, J. Singleton, V. Zapf, P. Manuel, I. I. Mazzin, Y. Li, H. O. Jeschke, R. Valentì, and R. Coldea, *Phys. Rev. B* **92**, 235119 (2015).
[26] S. M. Winter, K. Riedel, P. A. Maksimov, A. L. Chernyshev, A. Honecker, and R. Valenti, *Nature Comm.* **8**, 1152 (2017).

Acknowledgments

The authors acknowledge valuable discussions with J. van den Brink, R. Moessner, J. Knolle, I. Rouschata-
Author contributions

C.W., J.Z. and A.A. conducted the ESR and MWA studies and analyzed the data. M.R., A.I., and T.D. synthesized and characterized the samples. A.U.B.W. performed static magnetization measurements. V.K., B.B., A.A., and M.V. designed the project. All coauthors participated actively in the discussion and interpretation of experimental results and worked out the final concept of the paper. V.K., A.A. and M.V. wrote the paper with important contributions to all its parts from all other coauthors.

Competing financial interests

The authors declare no competing financial interests.
SUPPLEMENTARY INFORMATION

Details of the MWA measurements

All MWA measurements presented in the paper were performed upon sweeping the magnetic field from zero to 16 T at a given constant excitation frequency $\nu$. Data recorded during down sweeps showed similar results and are not shown. All data presented in this work have been obtained with the magnetic field applied within the $(ab)$-plane. We note that the major features discussed in the paper, in particular the absorption peak around a field of 7 T, were not observed in measurements with the external field perpendicular to the $ab$-plane.

Generally, in the framework of the linear response theory the MWA is determined by the imaginary part of the dynamic susceptibility at the frequency $\omega = 2\pi\nu$, the magnetic field $H$, and the wave vector $\vec{q} = 0$

$$\chi''(\omega, H) \propto \int_{-\infty}^{\infty} dt e^{i\omega t} \langle [S^+(t), S^-] \rangle_{T,H},$$

where $[S^+(t), S^-]$ is the transverse time-dependent spin correlation function with $S^+$ and $S^-$ being the raising and lowering spin operators, and the brackets $\langle \ldots \rangle_T$ denote the thermal average. Under certain instrumental conditions a real part of the dynamic susceptibility $\chi'(\omega)$ might be admixed to the measured signal causing a phase shift (dispersion) of the spectrum. Therefore we have carefully checked that the spectra recorded with the employed ESR setups are free of this artefact. In particular, the detection of the MWA signals with the vector network analyzer PNA-X and with the InSb bolometer yielded similar spectra. A representative comparison of the spectra is shown in Fig. S1. This ensures that all the spectra discussed in the paper represent the absorption signals, since the bolometer measures an incoming microwave power, which is proportional to the absorption due to the imaginary part of the dynamic spin susceptibility $\chi''(\omega)$, and is practically not sensitive to a possible admixture of $\chi'(\omega)$.

Fig. S2 shows a comparison of the resonance signals detected in our experiments at $T \ll T_N$, at frequencies larger than the minimum spin-wave gap energy $\Delta_{sw}(H) = 250$ GHz at $\mu_0 H = 7$ T, with the resonance branches $\nu$ vs. $H$ reported in Refs. [12–14]. Noting that the position of the resonance lines sensitively depends on the polarization of the microwaves, on the particular orientation of the external field $\vec{H}$ within the $ab$-plane of the crystal, as well as on an even small tilting of $\vec{H}$ out of the $ab$-plane, the agreement with the previous reports is very good. This emphasizes a similar excellent quality of the crystals investigated in the present work as compared to the samples from other sources studied in Refs. [12, 13].

![Fig. S1](image1.png)

**Fig. S1.** Comparison of the MWA spectra $S_0(H)$ detected by the vector network analyzer (PNA-X) and the InSb bolometer (ISB) at $T = 3$ K.

![Fig. S2](image2.png)

**Fig. S2.** Field dependence of the ESR modes at $T = 3$ K for $H \parallel ab$-plane (closed symbols) in comparison with the ESR branches "A, B, D, E, F" from Ref. [14] and "L1, M1" from Ref. [13] sketched by dot lines. Grey open circle at $H = 0$, $\nu = 620$ GHz, corresponds to the resonance mode from Ref. [12]. Light grey crosses denote the peaks in the spectrum which intensity rapidly fades by lowering the excitation frequency below the spin-wave gap value $\Delta_{sw}(\mu_0 H = 7$ T) $\approx 250$ GHz (1 meV) (cf. Fig. 1(a)).
FIG. S3. Contour plots presenting the color-coded MWA signal $S_d$ as a function of temperature and magnetic field. Red color indicates more absorption by the sample, and blue color indicates less absorption by the sample, relatively to the bias green color level. Measurements at three microwave frequencies (energies) are shown: (a) $\nu = 82$ GHz ($\sim 0.34$ meV); (b) $\nu = 273$ GHz ($\sim 1.13$ meV); (c) $\nu = 360$ GHz ($\sim 1.49$ meV).

Fig. S3 shows the $T-H$ color-coded maps of the signal at the detector $S_d$ for different excitation frequencies $\nu$. Data shown are obtained from recorded spectra by first subtracting the $S_d$ value at a magnetic field of 15.5 T, and afterwards by subtracting the spectrum measured at 30 K from spectra measured at other temperatures. Green color represents relative zero and ”red shift” (”blue shift”) of the color indicates increased (decreased) absorption of the microwaves by the sample.