High-field magnetic resonance of spinons and magnons in a triangular lattice \( S = 1/2 \) antiferromagnet \( \text{Cs}_2\text{CuCl}_4 \)

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The electron spin resonance doublet indicating the width of the two spinon continuum in a spin-\( 1/2 \) triangular-lattice Heisenberg antiferromagnet \( \text{Cs}_2\text{CuCl}_4 \) was studied in high magnetic field. The doublet was found to collapse in a magnetic field of a half of the saturation field. The collapse of the doublet occurs via vanishing of the high frequency component in a qualitative agreement with the theoretical prediction for the \( S = 1/2 \) chain. The field of the collapse is, however, much lower than expected for the \( S = 1/2 \) chain. This is proposed to be due to the destruction of frustration of interchain exchange bonds in a magnetic field, which restores the 2D character of this spin system. In the saturated phase the mode with the Larmor frequency and a much weaker mode downshifted for 119 GHz are observed. The weak mode is of exchange origin, it demonstrates a positive frequency shift at heating corresponding to the repulsion of magnons in the saturated phase.

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I. INTRODUCTION

Spin-1/2 Heisenberg antiferromagnet on a triangular lattice \( \text{Cs}_2\text{CuCl}_4 \) was extensively studied because of a remarkable two-spinon continuum of excitations, like that of the \( S = 1/2 \) antiferromagnetic chain. Other feature of this material is the delayed magnetic ordering at very low temperature and exotic field-induced phase transitions, which are due to weak interactions, while the dominant exchange interaction is frustrated. The aim of this work is to study the fine structure of the spinon continuum in a high magnetic field, including the transition to saturated phase.

In crystals of \( \text{Cs}_2\text{CuCl}_4 \) magnetic ions \( \text{Cu}^{2+} \) (\( S = 1/2 \)) are displayed in layers with a distorted triangular lattice, see Fig. 1. The 2D model Hamiltonian contains the following terms, see, e.g., Ref. 4:

\[
\hat{\mathcal{H}} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J' \sum_{\langle i,j' \rangle} \mathbf{S}_i \cdot \mathbf{S}_{j'} + \sum_{(i,k)} \mathbf{D}_{ik} \cdot \mathbf{S}_i \times \mathbf{S}_k , \tag{1}
\]

where \( J \) is the exchange integral for spins neighboring along \( b \)-direction, \( J' \) is the zig-zag interchain coupling, as shown in Fig. 1. Vectors \( \mathbf{D}_{ik} \) are parameters of Dzyaloshinsky-Moriya interaction. There are six different Dzyaloshinsky-Moriya vectors (\( \mathbf{D}_{1,2} \) and \( \mathbf{D}_{1',-4} \)) compatible with the symmetry of \( \text{Cs}_2\text{CuCl}_4 \). These vectors are shown schematically in Fig. 1 in the middle of each exchange bond. Vectors \( \mathbf{D}_{1,2} \) have nonzero \( a \)- and \( c \)-components of absolute values \( D_a \) and \( D_c \) and are oriented as shown in Fig. 1. Vectors \( \mathbf{D}_{1',-4} \) have nonzero components along all three crystallographic axes with absolute values \( D'_{a,b,c} \).

The main exchange integrals \( J, J' \) and the interplane exchange constant \( J'' \) were derived from the neutron scattering experiments in the saturated phase: \( J = 4.34(6) \) K, \( J' = 1.48(6) \) K, \( J'' = 0.22(3) \) K. Close values \( J = 4.7(2) \) K, and \( J' = 1.42(7) \) K were derived from the saturation field values and electron spin resonance (ESR) in the saturated phase. Several parameters of Dzyaloshinsky-Moriya interactions were extracted from experiments: inelastic neutron scattering gives \( D_a = 0.24 \) K, low-temperature ESR results in \( D_a = 0.23 \pm 0.05 \) K, \( D_c = 0.34 \pm 0.05 \) K, and high-temperature ESR linewidth gives \( D_a = 0.33 \) K, \( D_c = 0.36 \) K.

The observation of 1D excitation spectrum in this quasi 2D system is ascribed to the effective decoupling of spin chains because of the frustration of the antiferromagnetic exchange interactions \( J' \) on the diagonal bonds. This effective decoupling is supported by, e.g., numeri-
A feature of this compound is the uniform Dzyaloshinsky-Moriya interaction between the spins, neighboring along \( b \)-direction: vectors \( \mathbf{D}_{1,2} \) are equal in magnitude and direction for all bonds within a chain, in contrast to vectors \( \mathbf{D}'_{1-4} \) on diagonal bonds, which compose a staggered structure of a conventional Dzyaloshinsky-Moriya interaction. The uniform Dzyaloshinsky-Moriya interaction was predicted to modify the spectrum of excitations in a 1D \( S=1/2 \) chain. This interaction should cause a shift of the continuum in \( \mathbf{q} \)-space by a vector \( \mathbf{q}_{\text{DM}} = D J/\hbar \). As a result, in a magnetic field \( \mathbf{H} \parallel \mathbf{D} \), the ESR line should split into a doublet. The frequencies of the doublet components are at the upper and lower boundaries of the initial (i.e. unshifted) continuum at the wave vector \( \mathbf{q}_{\text{DM}} \), see Refs. 12,13. At the same time, ESR signal should not split at the orthogonal orientation of magnetic field. In this case a gap of the ESR absorption in zero field should open. The doublet is marking the width of the continuum and appears due to the fractionalized character of excitations and to uniform Dzyaloshinsky-Moriya interaction.

The ESR doublet arising at \( \mathbf{H} \parallel \mathbf{D} \) and merging into a single line at \( \mathbf{H} \perp \mathbf{D} \) was indeed observed experimentally in the spin-liquid phase of \( \text{Cs}_2\text{CuCl}_4 \) in Refs. 7,8.

In the preceding ESR paper describing high-field experiments, magnetic field was oriented parallel to \( b \)-axis, because the exchange parameters may be derived most accurately by use of this orientation. However, at \( \mathbf{H} \parallel b \) magnetic field is perpendicular to \( \mathbf{D} \) and the spinon doublet does not appear. Therefore, the high-field evolution of the ESR spinon doublet (i.e. of the width of the spinon continuum near the Brillouin zone center) was not addressed there. The aim of the present work is to follow experimentally the evolution of the ESR doublet in a high magnetic field, i.e at frequencies of the exchange range. A vanishing of the doublet is expected in a high field, because of the suppressing of quantum fluctuations. In particular, this should close the width of the spinon continuum at saturation (see theory, e.g., Ref. [14]). We indeed detect the vanishing of the doublet at \( \mathbf{H} \parallel a \). It occurs via the cease of the high frequency component of the doublet. Besides, we observe the transformation of the spinon ESR response into two ESR modes in the saturated phase. One of these modes originates from the magnon branch with a dispersion along \( c \)-axis. This mode shows an effect of the frequency shift at heating, indicating a repulsion of magnons.

II. EXPERIMENT

The preceding investigation shows, that the spinon doublet is most clearly pronounced at lowest temperatures and that at the frequency above the exchange value \( J/h \simeq 80 \text{ GHz} \) it is not affected by cooling below \( T_N = 0.62 \text{ K} \). The change of the doublet to an antiferromagnetic resonance spectrum was observed only below 40 GHz. This conservation of a spin-liquid spectrum at high energy range, and arising of spin-wave modes at low energy is typical for systems with a delayed ordering occurring far below Curie-Weiss temperature (see, e.g., Refs. [15,16]). For the above reason, to measure the high-field evolution of ESR signal we choose the temperature of about 0.5 K and the frequency above 60 GHz. Here the components of the doublet are well separated and at the same time are not affected by the ordering. We align the magnetic field along the \( a \)-axis, because at this orientation the doublet is well seen and the spin structure evolves gradually to saturation without phase transitions even in the ordered phase. We used the crystals of \( \text{Cs}_2\text{CuCl}_4 \) from the same batch as in Refs. [7,8].

Experiments were performed using a set of ESR spectrometers, operating with superconducting 12 T magnet, combined with a \( ^3\text{He} \) cryo-insert, providing low temperature down to 0.45 K. A small amount of powder

![FIG. 2: Examples of ESR lines of \( \text{Cs}_2\text{CuCl}_4 \) at \( \mathbf{H} \parallel a \) at different frequencies. The zoom for the area within a circle is 6-fold for vertical scale and 3-fold for horizontal scale.](image-url)
of 2,2-diphenyl-1-picrylhydrazyl (known as DPPH) was employed as a standard $g = 2.00$ marker for the field. Backward wave oscillators were microwave sources, covering the range 60–350 GHz. The microwave units of two types were used for recording the resonance absorption of microwaves. In the first unit cylindrical multi-mode resonators were used as plug-in components in a transmission microwave circuit. The second unit is a narrowed waveguide with a diaphragm, also used in a transmission mode. In case of a properly tuned resonator we observe the diminishing of the transmission, proportional to the imaginary part of the susceptibility of the sample. The ESR line of a conventional paramagnet recorded in this way should have a Lorentzian shape. Unfortunately, for frequencies above 200 GHz the spectrum of eigenfrequencies of the cavity is too dense and proper tuning is difficult. The waveguide doesn’t require frequency tuning, but in this case the change of transmission is a superposition of the real ($\chi'$) and imaginary ($\chi''$) parts of microwave susceptibility (see, e.g. Ref. [1]).

![Graph showing transmission vs. magnetic field for different temperatures](image)

**FIG. 3**: (Color online) 142 GHz ESR records at $H \parallel a$ in Cs$_2$CuCl$_4$ at several temperatures.

It should be noted, that at the frequency above 140 GHz the samples of a size of about 2 mm have strongly distorted (indented) ESR line because of parasitic field-dependent resonances, which arise due to the large dynamic susceptibility $\chi'$ of the sample near the resonance field. The susceptibilities $\chi'$, $\chi''$ change strongly and not monotonously within an interval of several linewidths $\Delta H$ on both sides of the resonance field $H_0$. For a typical paramagnet (see, e.g., Ref. [18]) the susceptibility $\chi'$ is negative in an interval below the resonance field, then it takes positive value, reaching a maximum at $H = H_0 \approx \frac{1}{2} \Delta H$, and then gradually drops to zero. Thus, the large positive values of $\chi'$ occur twice during a sweep of the field across the right wing of the resonance curve. The large value of $\chi'$ may result in eddy current resonances in the dielectric sample at field values, when a half of the electromagnetic wave length is comparable to the sample size or fractional value of the sample size. In this way, for a high frequency, large sample and high susceptibility, several eddy current resonances may arise in the field interval, where the real part of the susceptibility is rising and the same resonances should again occur in the field range, where the susceptibility is falling. In this case the ESR lineshape appears to be distorted by indenting via parasitic resonances. To avoid this parasitic effect, one has to use a method of transmission of plane electromagnetic waves through the sample, having a thin plate shape. Here the eddy current resonances are fixed as interference pattern of plane waves. Another way is to diminish the sample size far below the half of the length of the electromagnetic wave within the sample. We used the samples of the size below 0.5 mm for recording strong ESR signals and samples with the size of about 2 mm to detect the weak ESR line, which arises above the saturation field. Besides, a test for parasitic resonances may be performed in the paramagnetic phase at $T > 10$ K, when the imaginary part of the ESR susceptibility is surely a Lorentzian function of the magnetic field, and the real part is also a known function of field, see, e.g., Ref. [18]. The manipulation with the sample size and the test by means of the paramagnetic resonance enables one to avoid parasitic eddy current resonances and to fix the intrinsic lineshape in the range below 250 GHz. At higher frequencies ESR lines appeared indented, this resulted in a higher error of the measurement of resonance field.

### III. EXPERIMENTAL RESULTS

The evolution of the ESR lineshape with changing frequency at $H \parallel a$ is shown in Fig. 2. Here the ESR records taken by resonator unit in the range 70–150 GHz and by waveguide unit at 245 GHz are shown. We see, that the low-field (i.e. high-frequency) component of the doublet loses intensity with the increase of the magnetic field. For frequencies above 140 GHz the doublet disappears completely, and only a single ESR line with the paramagnetic resonance frequency $f_0 = g_\mu_B H/(2\pi\hbar)$, $g_a = 2.20$ is observed below the saturation field 8.44 T. At the further increase of the magnetic field we continue to observe a strong ESR line with the frequency $f_0 = g_\mu_B H/(2\pi\hbar)$. Besides, we see a much weaker ESR line in the magnetic field above 8 T (mode "B"), see upper curve in Fig. 2. The ratio of integral intensity of the weaker mode B to the intensity of the $f_0$ mode is about 0.015. The 245 GHz curve presents an example of the parasitic indentation of the intensive resonance for a sample, which is oversized for the high microwave susceptibility of $f_0$-mode, but provides a good sensitivity for a weak mode B.

Fig. 3 demonstrates records of 142 GHz ESR at several temperatures. This records, performed by a resonator...
unit in the middle part of the frequency range present both intensive and weak modes for the same sample without parasitic distortions. The temperature evolution of the intensive line shows vanishing of the doublet component \( A' \) in the almost collapsed doublet by heating. The weak line appearing above the saturation field also disappears at heating.

The transformation of the doublet into a single ESR line with the increase of the magnetic field is illustrated in Fig. 4, here the field dependence of the shift of doublet components with respect to \( f_0 \) is shown in the upper panel. Data presented here are taken in the frequency range 60–200 GHz at \( T = 0.5 \) K. The lower panel shows the ratio of amplitude of the upper component \( u_A \) to the lower component amplitude \( u_{A'} \). The collapse of the doublet occurs in the magnetic field of 4.0 T, which constitutes approximately \( 0.5 H_{sat} \). The frequency-field dependence of all modes at \( T = 0.5 \) K is presented in Fig. 5.

The weaker mode \( B \), arising above the saturation field, was observed also at \( H \parallel b \) in Ref. 6. We study here the temperature dependence of this mode. The orientation of the field \( H \parallel b \) is selected because the theory has maximal accuracy at this direction of the field. The resonance field of the mode \( B \) exhibits a shift to lower fields at heating, as demonstrated in Fig. 6. The temperature dependence of the resonance field and of the linewidth are shown in Fig. 7.

FIG. 4: (Color online) Upper panel: shift of the resonance frequencies of doublet components \( A \) and \( A' \) relative to paramagnetic resonance. Crossed circles are 35 GHz data for the spin-liquid phase at \( T = 1.0 \) K. Other symbols correspond to \( T=0.5 \) K. Solid line presents calculation of the continuum boundaries at the wavevector \( q_D \) following Ref. 14. Lower panel: ratio of amplitudes of the doublet components \( u_{A'}/u_A \) vs resonance field of \( A \)-component. Dashed lines in both panels are guides to the eyes.

IV. DISCUSSION

The spinon continuum and the related ESR doublet observed in Cs\(_2\)CuCl\(_4\) are consequences of quantum fluctuations in a spin system, which remains paramagnetic (spin-liquid) at temperatures far below the Curie-Weiss temperature. Magnetization should suppress zero-point fluctuations and, hence, the doublet should be transformed in a single ESR line with the Larmor frequency, at least in the saturated phase. The application (see Appendix A) of the theory of a Heisenberg \( S=1/2 \) antiferromagnetic chain predicts a collapse of the spinon continuum width (and, hence, of the considered doublet) at the saturation field, see solid line in the upper panel of Fig. 4. The exchange integral \( J \) and Dzyaloshinskii-Moriya parameter \( D_a \) of Cs\(_2\)CuCl\(_4\) was used for this calculation. Note, that the saturation field 6.3 T calculated in 1D model is lower than the more realistic 2D value of 8.44 T.

Our observations confirm that the collapse of the dou-
A doublet really occurs. However, the doublet does not survive till the saturation field, but collapses in the field of about 0.5\( H_{sat} \). This discrepancy between the observed behavior of the spinon doublet and the theory of a spin chain may be attributed, probably, to the cease of the frustration of the exchange coupling between chains in Cs\(_2\)CuCl\(_4\) in a magnetic field. The frustration of interchange coupling takes place in zero field, when the antiferromagnetic correlation of neighboring spins within the chain prevails.\(^{10}\) The antiferromagnetic correlation changes to a ferromagnetic one in a strong magnetic field, thus, the interaction between the chains should be restored and 1D consideration becomes inapplicable to Cs\(_2\)CuCl\(_4\) in a strong field.

The vanishing of the upper component of the doublet is qualitatively consistent with the theoretical investigation of the spectral density of two spinon continuum of the 1D \( S = 1/2 \) Heisenberg antiferromagnet in a magnetic field.\(^{20}\) In this theoretical study the intensity at the upper boundary of the spinon continuum of the transverse spin oscillations is shown to drop in process of magnetization (see Fig. 3 of Ref.\(^ {20} \)).

The experiment in the field, exceeding the saturation point \( \mu_0 H_{sat} = 8.44 \) T, shows the downshift 119±2 GHz of the frequency of the weak mode B with respect to the Larmor frequency of mode A. The appearance of a downshifted mode is in a good correspondence with the theory of the spin wave excitations of the saturated phase, given in the Supplement material of Ref.\(^ {6} \). This theory predicts approximately the same ESR frequencies for the field both perpendicular and tilted to vector \( \mathbf{D} \):

\[
2\pi \hbar f_{ESR} = g_{a,b,c} \mu_B H_{a,b,c} - 4J' + O(D/J)^2. \tag{2}
\]

The same shift 119 GHz was observed for the mode B for another orientation of the magnetic field \( \mathbf{H} \parallel b \) in Ref.\(^ {6} \). Thus, the theoretical prediction on the approximately isotropic character of the shift of mode B corresponds well to the theory. It should be noted, that the mode B, observed here by ESR method, i.e. at the Brillouin zone center, is the same excitation, as observed by inelastic neutron scattering at the boundary of the "exchange" Brillouin zone (\( k_c = \pi/c \)). Indeed, in Ref.\(^ {5} \) the Brillouin zone was considered in the exchange approximation with periods \( \delta_{1,2} \). However, the structure composed by vectors \( \mathbf{D}' \) has a doubled period in \( c \)-direction in comparison with the exchange structure, see Fig.\(^ {1} \). The period doubling results in the folding of the Brillouin zone. Thus, excitations, positioned at the boundary of...
the zone in the exchange approximation, appear in the center of the zone (see Fig. 1 in Ref. [8]). The weak but nonzero ESR intensity of this mode is attributed, thus, to the Dzyaloshinsky-Moriya interaction and would be zero in the exchange approximation (see theory in Ref. [8]).

The negative shift of the resonance field of mode B, observed at heating, means the enlarging of the eigen frequency of magnons at excitation of additional magnons. This may be treated as a consequence of a repulsive interaction of magnons. The repulsion of magnons is natural for fully polarized antiferromagnetic system, because here two flipped spins show a mutual repulsion. The mode B practically disappears above 2 K. This is also natural as the dispersion in \( k_- \)-direction is provided by the exchange \( J' = 1.45 \) K. This dispersion determines the frequency of the ESR mode B. Thus, the temperature, higher than \( J' \), should smear this resonance mode, as seen in the experiment.

The nonlinear spin-wave calculations within \( J-J' \) Heisenberg model in the saturated phase\(^{22}\) give the following expression for the temperature-dependent energy shift at the wavevector \( q = (0, 2\pi/3) \), corresponding to the frequency of ESR mode B:

\[
\delta \varepsilon = 8 J' \sum_{k} \left( 1 - \cos \frac{k_{\parallel}}{2} \cos \frac{\sqrt{3} k_{\perp}}{2} \right) \exp(\frac{\varepsilon(k)}{T}) + 1 \tag{3}
\]

Here the wavevector is measured in units of reciprocal periods of the 2D lattice with the translations \( \delta_{1,2} \) in Fig. 1. The magnon spectrum \( \varepsilon(k) \) is described by the relation 26 of Supplemental material of Ref. [6].

The result of the calculation after relation (3) is shown in the upper panel of Fig. 7. This calculation is made for \( \mu_B H = 11 \) T and exchange parameters \( J \) and \( J' \) for \( \text{Cs}_2\text{CuCl}_4 \) from Ref. [8]. The result of the experiment corresponds well to the theory both in the sign and the value of the shift.

## V. CONCLUSION

The evolution of the electron spin resonance spectrum in the frequency range above the exchange frequency \( J/(2\pi h) \) was studied in the \( S = 1/2 \) antiferromagnet on the distorted triangular lattice \( \text{Cs}_2\text{CuCl}_4 \). The doublet of resonance lines, marking the boundaries of the spinon continuum was found to collapse in the field of about a half of the saturation field. The collapse proceeds via vanishing of the upper frequency component of the doublet. This scenario of the collapse of the doublet agrees qualitatively with the evolution of the spinon continuum of spin \( S = 1/2 \) Heisenberg antiferromagnetic chain\(^{14,20} \). Above the saturation field, a much weaker mode, downshifted for 119 GHz from the Larmor frequency is observed. This shift and the weak intensity of this mode correspond well to the theoretical consideration of spin waves in the saturated phase. The temperature dependence of the resonance field of the weaker mode indicates the repulsive interaction of magnons in the saturated antiferromagnet and is well explained within the spin wave formalism with anharmonic terms.

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## Appendix A: ESR frequencies of the spin chain in high fields

Under the action of the applied magnetic field continua of spin fluctuations of different polarization become different. The spectra of continua of transverse spin fluctuations \( S_{+-} \) and \( S_{-+} \) are responsible for ESR absorption\(^{21} \). The upper and lower boundaries of these continua for \( S = 1/2 \) Heisenberg antiferromagnetic chain in presence of a magnetic field are calculated by Müller et al. These data are summarized in Table II of Ref. [14]. As described in Sec. I, to calculate the upper and lower boundary frequencies at \( q = 0 \) for a spin chain with the uniform Dzyaloshinsky-Moriya interaction we use the results of Ref. [14], taking the boundary frequencies at \( q = q_{\text{DM}} \) for the boundaries with non-vanishing spectral weight.

In this way we obtain the following frequency-field dependencies for spin-resonance absorption at \( H \parallel D \):

\[
2\pi h f_1 = J R(h) \left[ \cos \left( \frac{q_{\text{DM}}}{2} \right) + \frac{\pi m(h)}{2} \right] - J h,
\]

\[
\tag{A1}
2\pi h f_2 = J R(h) \left[ \sin \left( \frac{q_{\text{DM}}}{2} \right) \cos \left( \frac{\pi}{2} - \frac{q_{\text{DM}}}{2} \right) - \frac{\pi m(h)}{2} \right],
\]

\[
\tag{A2}
2\pi h f_3 = J R(h) \left[ \sin \left( \frac{\pi}{2} + \frac{q_{\text{DM}}}{2} \right) \cos \left( \frac{\pi}{2} + \frac{q_{\text{DM}}}{2} - \frac{\pi m(h)}{2} \right) \right],
\]

\[
\tag{A3}
R(h) = \left( \pi + h(1 - \frac{\pi}{2}) \right) \text{ is the field-dependent renormalization prefactor and } m(h) \text{ is the reduced magnetization given by}^{14}
\]

\[
m(h) = \frac{1}{\pi} \arcsin \left( \frac{1}{1 - \pi/2 + \pi/h} \right). \tag{A4}
\]

Domains of these functions in \( q \) and \( H \) are chosen to avoid negative values of frequencies. Using equations
we get the ESR frequencies shown in Fig. 4 for the $S = 1/2$ spin chain with uniform Dzyaloshinsky-Moriya interaction in the so-called Müller ansatz approximation: mode $f_1$ is given by the lower boundary of $S_+ -$ continuum and $f_{2,3}$ are given by the lower boundary of $S_+ +$ continuum. In case of low field $h \ll 1$ these equations transform into corresponding relations of Refs. 7, 12. Modes $f_2$ and $f_3$ correspond to $A'$ and $A$ in Fig. 4 and mode $f_1$ is relevant only for small magnetic fields which are out of range of the present study.

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