High-field magnetic resonance of spinons and magnons in the triangular lattice $S = \frac{1}{2}$ antiferromagnet Cs_2CuCl_4

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The electron spin resonance doublet indicating the width of the two-spinon continuum in a spin-$\frac{1}{2}$ triangular lattice Heisenberg antiferromagnet Cs_2CuCl_4 was studied in high magnetic field. The doublet was found to collapse in a magnetic field of one-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 $S = \frac{1}{2}$ chain. The field of the collapse is, however, much lower than expected for the $S = \frac{1}{2}$ 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

The Heisenberg antiferromagnet on a triangular lattice is a challenging object both for theoreticians and experimentalists, because of chiral and noncollinear ordering, also because of unusual phase transitions induced by magnetic field, which reveal phases, stabilized by order-by-disorder mechanism. Besides, the $S = \frac{1}{2}$ representatives of this family of magnets attract attention by possible spin-liquid behavior with an absence of the magnetic ordering even at absolute zero. For a description of these phenomena, see, e.g., review articles [1,2]. Numerous experimental realizations of this model have demonstrated that the search for unusual magnetism in these systems was not in vain. One of the representatives of this family is the spin $S = \frac{1}{2}$ Heisenberg antiferromagnet on a triangular lattice Cs_2CuCl_4. It was extensively studied because of different manifestations of effects related to frustration and quantum fluctuations. One of these effects is a delay of the magnetic ordering from the Curie-Weiss temperature $T_{CW} = 4$ K till a much lower temperature $T_N = 0.6$ K. This compound exhibits exotic field-induced phase transitions [3], which occur due to the disturbance of a fine balance of weak interactions, while the dominant exchange interaction is frustrated [4]. The strong delay of the ordering enables one to study static and dynamic properties in a temperature range $T_N < T < T_{CW}$, where a spin-liquid-like phase is formed. In particular, a remarkable two-spinon continuum of excitations, like that of the $S = \frac{1}{2}$ antiferromagnetic chain, was found in this quasi-2D spin structure [5,6].

The surprising observation of a 1D excitation spectrum in the quasi-2D system is ascribed to the effective decoupling of spin chains because of the frustration of the antiferromagnetic exchange bonds J' at the lateral bonds of the triangular structure. This effective decoupling is confirmed by, e.g., numerical simulations [7] and an analytical approach [8].

Further interest in this system was stimulated by the so-called "uniform" Dzyaloshinsky-Moriya interaction, which is a feature of this compound. The uniform Dzyaloshinsky-Moriya interaction causes classical spins to form a spiral structure in contrast to a canted antiferromagnet in the case of a conventional "staggered" Dzyaloshinsky-Moriya interaction [9,10]. For an $S = \frac{1}{2}$ antiferromagnetic chain, which has a quantum-disordered ground state, the uniform Dzyaloshinsky-Moriya interaction affects the spinon modes of excitations and their spectrum in the low-energy range [11,12]. The two-spinon continuum was predicted to be modified via a shift in **q** space by a vector $q_{DM} = \frac{D}{J}\frac{1}{b}$. Here J is the main exchange integral in the spin chain and $\mathbf{D}$ is the vector parameter of the uniform Dzyaloshinsky-Moriya interaction. As a result, in a magnetic field $\mathbf{H} \parallel \mathbf{D}$, the line of the electron spin resonance (ESR) 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 q_{DM}; see Refs. [11,12]. At the same time, the ESR signal should not split at the orthogonal orientation of the 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 a 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 Cs_2CuCl_4 in Refs. [13,14].

The aim of this work is to study the above doublet of the fine structure of the spinon continuum in a high magnetic field, including the transition to the saturated phase. This implies ESR 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., in Ref. [15]). By use of the multifrequency ESR we indeed detect the vanishing of the doublet. It occurs via the ceasing

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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 the c axis. This mode shows an effect of the frequency shift at the increase of temperature, indicating a repulsion of magnons.

II. EXPERIMENT

Experiments were performed using a set of ESR spectrometers, operating with a superconducting 12 T magnet, combined with a ^{3}He cryo-insert, providing low temperature down to 0.45 K. A small amount of powder 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 multimode 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 the 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 does not require frequency tuning, but in this case the change of transmission is a superposition of the real (χ') and imaginary (χ'') parts of microwave susceptibility (see, e.g., Ref. [16]).

It should be noted that at the frequency above 140 GHz the samples of a size of about 2 mm have a strongly distorted (indented) ESR line because of parasitic field-dependent resonances, which arise due to the large dynamic susceptibility χ' of the sample near the resonance field. The susceptibilities χ', χ'' change strongly and not monotonically within an interval of several linewidths ΔH on both sides of the resonance field H_0. For a typical paramagnet (see, e.g., Ref. [17]) the susceptibility χ' is negative in an interval below the resonance field, then it takes positive value, reaching a maximum at $H - H_0 \simeq \frac{1}{2}\Delta H$, and then gradually drops to zero. Thus, the large positive values of χ' occur twice during a sweep of the field across the right wing of the resonance curve. The large value of χ' may result in electrodynamic resonances in the dielectric sample at field values, when one-half of the electromagnetic wavelength 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 electrodynamic 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 line shape 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 [18]. Here the electrodynamic resonances are fixed as interference patterns of plane waves. Another way is to diminish the sample size far below one-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. [17]. The manipulation with the sample size and the test by means of the paramagnetic resonance enables one to avoid parasitic electrodynamic resonances and to fix the intrinsic line shape 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.

For observations of the doublet in the high-frequency range we used the temperature of about 0.5 K, because the resolution of the doublet is most clearly seen at most low temperature. As shown in Ref. [14], the ordering at a close temperature $T_N = 0.62$ K does not prevent the observation of the spinon doublet at frequencies above the exchange value $J/\hbar \simeq 80$ GHz, because the change of the doublet to an antiferromagnetic resonance spectrum was found only below 40 GHz. This conservation of a spin-liquid spectrum at high energy range, and a change to spin-wave modes at low energy with cooling through T_N, is typical for low-dimensional systems with a delayed ordering occurring far below Curie-Weiss temperature (see, e.g., Refs. [19,20]). 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.

The control of the temperature of the microwave unit in the temperature range 0.5–4 K was performed by use of a heater placed on the microwave unit and/or by regulation of ^{3}He pumping. The temperature was measured by a RuO_2 thermometer placed on a microwave unit. To avoid the overheating of the sample by microwaves we used test records of ESR lines at different levels of microwave power, as described in Ref. [14].

We used the crystals of Cs_2CuCl_4 from the same batch as in Refs. [13,14,21]. The samples were oriented with respect to the external field with an accuracy of 3°.

III. EXPERIMENTAL RESULTS

The evolution of the ESR line shape with changing frequency at $\mathbf{H} \parallel a$ is shown in Fig. 1 for $T = 0.5$ K. Here the ESR records taken for a sample with the weight 1.8 mg in the resonator unit in the range 70–150 GHz are shown. The record taken at 245 GHz (upper curve in Fig. 1) is for the sample of the weight 5.0 mg in a waveguide unit. 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 $f_0 = g_a\mu_B\mu_0 H/(2\pi\hbar)$, $g_a = 2.20$ is observed almost till the saturation field 8.44 T (this value is given after Ref. [3]). At the further increase of the magnetic field we continue to observe a strong ESR line with the frequency f_0. Besides, we see a much weaker ESR line in the magnetic field above 8 T (mode B); see upper curve in Fig. 1. The ratio of integral intensity of the weaker mode B to the intensity of the f_0 mode is about

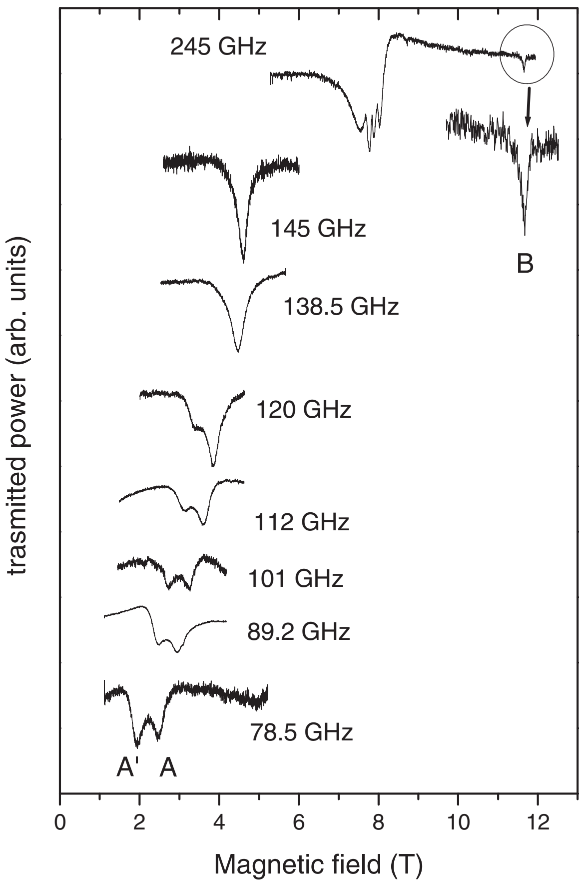

FIG. 1. Examples of ESR lines of Cs_2CuCl_4 at $\mathbf{H} \parallel a$, $T = 0.5$ K at different frequencies. The zoom for the area within a circle is 6-fold for vertical scale and 3-fold for horizontal scale.

0.015. The 245 GHz curve presents an example of the parasitic indention of the intensive resonance, as described in Sec. II. Thus, the sample used for this record appeared to be oversized for the high microwave susceptibility of the f_0 mode, but has an optimal size for recording a weak mode B.

Figure 2 demonstrates records of 142 GHz ESR at several temperatures. These records, performed by a resonator unit in

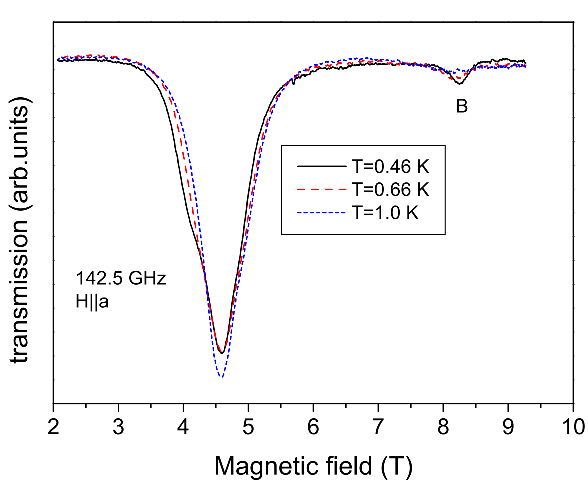

FIG. 2. (Color online) 142 GHz ESR records at $\mathbf{H} \parallel a$ in Cs_2CuCl_4 at several temperatures.

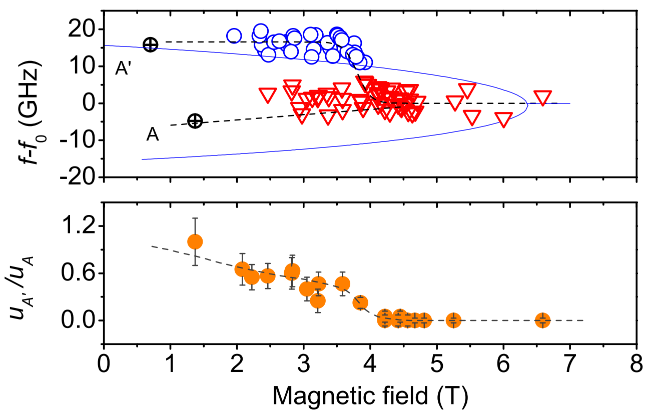

FIG. 3. (Color online) Upper panel: Shift of the resonance frequencies of doublet components A and A′ relative to paramagnetic resonance. Crossed circles are 35 GHz data [14] for the spin-liquid phase at $T = 1.0$ K. Other symbols correspond to $T = 0.5$ K. Solid line presents calculation of the continuum boundaries at the wave vector q_D following Ref. [15]. 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.

the middle part of the frequency range for a 6.0 mg sample, 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 at increasing temperature. 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. 3; here the field dependence of the shift of the resonance fields of doublet components with respect to ESR field of Larmor precession with the frequency 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 $0.5H_{sat}$. The overview of the frequency-field dependencies of all modes at $T = 0.5$ K is presented in Fig. 4.

The weaker mode B, arising above the saturation field, was observed also at $\mathbf{H} \parallel b$ in Ref. [21]. We study here the temperature dependence of this mode. The orientation of the field $\mathbf{H} \parallel b$ is selected because the theory [21] has maximal accuracy at this direction of the field. The resonance lines of mode B taken at different temperatures are shown in Fig. 5. One can see that the resonance field is shifted towards lower field at the increase of temperature. The corresponding temperature dependencies of the shift of the resonance field with respect to the resonance field H_{B0} (measured at $T = 0.5$ K) and of the linewidth are shown in Fig. 6.

IV. DISCUSSION

In crystals of Cs_2CuCl_4 magnetic ions Cu^{2+} ($S = \frac{1}{2}$) are displayed in layers with a distorted triangular lattice; see Fig. 7. The 2D model Hamiltonian contains the following terms (see,

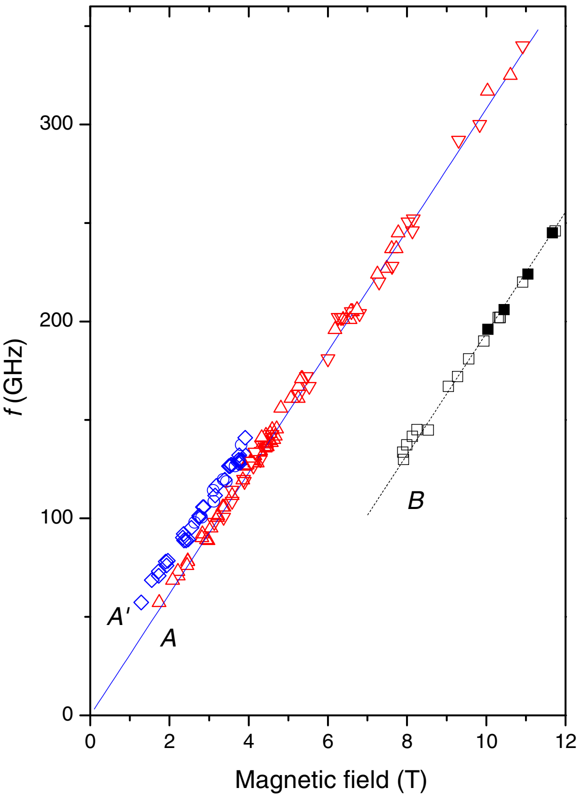

FIG. 4. (Color online) Frequency-field diagram of ESR response in Cs_2CuCl_4 with magnetic field along the a axis ($T = 0.5$ K). Data presented by symbols $\triangle$, $\diamond$, $\blacksquare$ are taken by use of the waveguide unit, $\triangledown$, $\bigcirc$, $\square$ by resonator unit. The solid line presents the Larmor frequency f_0 (see text). Dashed line is the Larmor frequency downshifted for 119 GHz.

e.g., Ref. [4]):

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

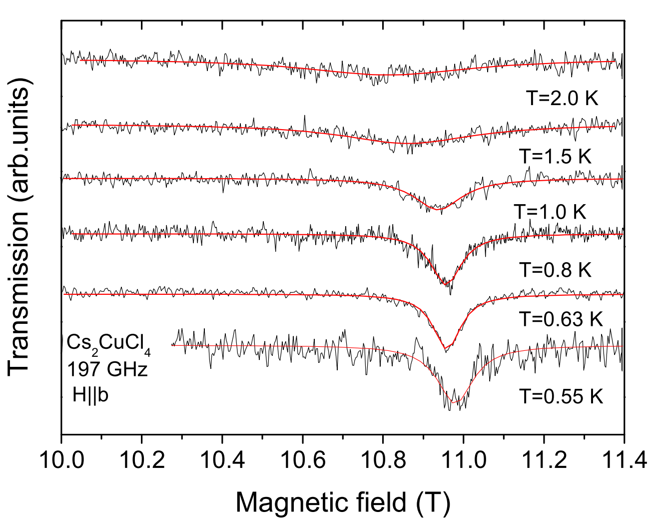

FIG. 5. (Color online) Temperature evolution of line B at the frequency 197 GHz and $\mathbf{H} \parallel b$. Solid lines are Lorenz fits for experimental curves.

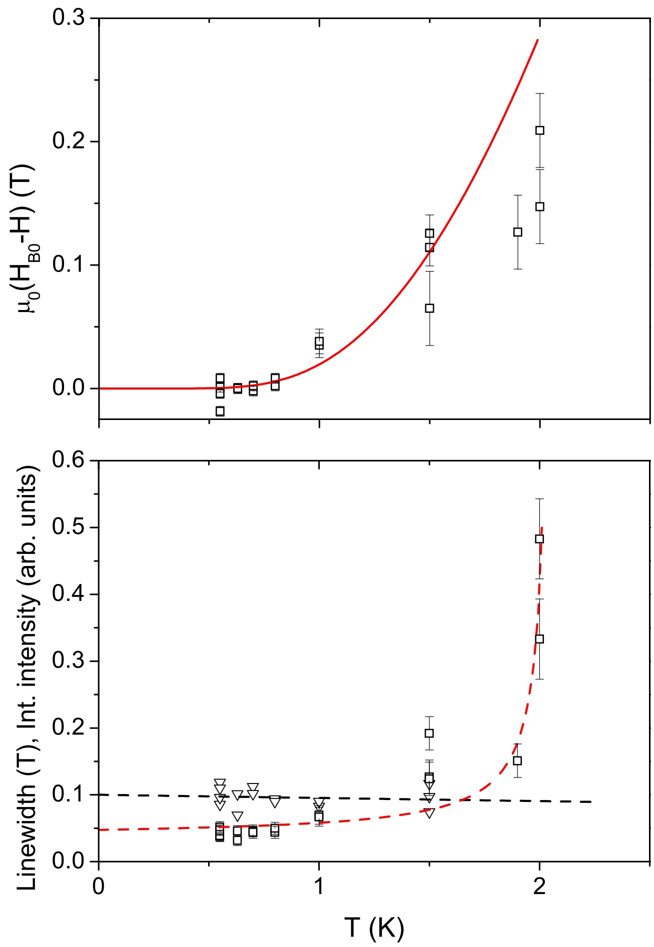

FIG. 6. (Color online) Upper panel: Temperature-induced shift of the resonance fields of line B at the frequency 197 GHz, $\mathbf{H} \parallel b$. The reference field $\mu_0 H_{B0} = 11$ T is the ESR field for mode B at $T = 0.5$ K. Solid line presents the theoretical calculation according to relation (3). Lower panel: Temperature dependence of the integral intensity (triangles) and linewidth (squares) of mode B at 197 GHz. Dashed lines are guides to the eyes.

here J is the exchange integral for spins neighboring along the b direction, and J' is the zigzag interchain coupling, as shown in Fig. 7. Vectors $\mathbf{D}_{ik}$ are parameters of the Dzyaloshinsky-Moriya interaction. There are six different Dzyaloshinsky-Moriya vectors ($\mathbf{D}_{1,2}$ and $\mathbf{D}'_{1-4}$) compatible with the symmetry of Cs_2CuCl_4 [4]. These vectors are shown schematically in Fig. 7 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. 7. 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 [22]: $J = 4.34(6)$ 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 value [3] and electron spin resonance (ESR) in the saturated phase [21]. Several parameters of Dzyaloshinsky-Moriya interactions were extracted from experiments: inelastic neutron scattering [22] gives $D'_a = 0.24$ K, low-temperature ESR [13,14] results in $D_a = 0.23 \pm 0.05$

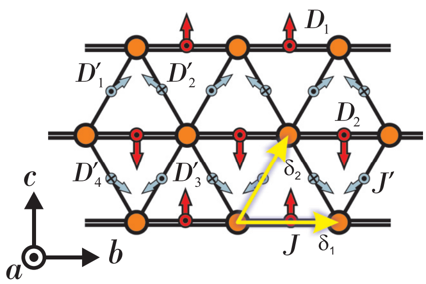

FIG. 7. (Color online) Sketch of the exchange paths of Cs_2CuCl_4 within a bc layer. Large circles mark Cu ions. J and J' are exchange integrals for two kinds of bonds. $\mathbf{D}_{1,2}$ and $\mathbf{D}'_{1-4}$ are Dzyaloshinsky-Moriya vectors according to Ref. [4]. Out-of-plane components of vectors are marked by points and crosses. Translations $\delta_{1,2}$ are periods for exchange bond structure.

K, $D_c = 0.34 \pm 0.05$ K, and high-temperature ESR linewidth [23] gives $D_a = 0.33$ K, $D_c = 0.36$ K.

As mentioned in Sec. I, a feature of this compound is the uniform Dzyaloshinsky-Moriya interaction between the spins, neighboring along the 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 spinon continuum and the related ESR doublet observed in Cs_2CuCl_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 the Appendix) of the theory of a Heisenberg $S = \frac{1}{2}$ antiferromagnetic chain [15] 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. 3. The exchange integral J and Dzyaloshinsky-Moriya parameter D_a of Cs_2CuCl_4 was used for this calculation. Note that the saturation field 6.3 T calculated in 1D model is lower than the real value of 8.44 T.

Our observations confirm that the collapse of the doublet really occurs. However, the doublet does not survive till the saturation field, but collapses in the field of about $0.5H_{\rm sat}$. This discrepancy between the observed behavior of the spinon doublet and the theory of a spin chain may be attributed, probably, to the ceasing of the frustration of the exchange coupling between chains in Cs_2CuCl_4 in a magnetic field. Indeed, the frustration of interchange coupling takes place in zero field, when the antiferromagnetic correlation of neighboring spins within the chain prevails [7]. 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_2CuCl_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 the two-spinon continuum of the 1D $S = \frac{1}{2}$ Heisenberg antiferromagnet in a magnetic field [24]. In this theoretical study the intensity at the upper boundary of the spinon continuum of the transverse spin oscillations is shown to drop in the process of magnetization (see Fig. 3 of Ref. [24]).

The experiment in the field, exceeding the saturation point $\mu_0 H^a_{\rm sat} = 8.44$ 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 Supplemental Material of Ref. [21]. This consideration implies a spin-wave theory used for a fully polarized phase of Cs_2CuCl_4 for which the ground state is exactly known and the excitation spectrum may be strictly calculated. This theory predicts approximately the same ESR frequencies for the field both perpendicular and tilted to vector $\mathbf{D}$:

$$2\pi\hbar f_{\rm ESR} = g_{a,b,c}\mu_B\mu_0 H_{a,b,c} - 4J' + O(D/J)^2. \quad (2)$$

The same shift 119 GHz was observed for the mode B for another orientation of the magnetic field $\mathbf{H} \parallel b$ in Ref. [21]. 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 the ESR method, i.e., at the Brillouin zone center, is the same excitation as observed by inelastic neutron scattering [22] at the boundary of the "exchange" Brillouin zone ($k_c = 2\pi/c$). Indeed, in Ref. [22] 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 the c direction in comparison with the exchange structure; see Fig. 7. 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. [21]). 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. [21]).

The negative shift of the resonance field of mode B, observed at heating, means the enlarging of the eigenfrequency 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 in a polarized antiferromagnet two flipped spins show a mutual repulsion. The mode B practically disappears above 2 K. This is also natural as the dispersion in the k_c 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 the J-J' Heisenberg model in the saturated phase [25] give the following expression for the temperature-dependent energy shift at the wave vector $\mathbf{k} = (0, 2\pi/\sqrt{3})$, corresponding to the frequency of ESR mode B:

$$\delta\varepsilon = \frac{1}{N} 8J' \sum_{\mathbf{k}} \frac{1 - \cos\frac{k_x}{2}\cos\frac{\sqrt{3}k_y}{2}}{\exp\left(\frac{\varepsilon(\mathbf{k})}{k_B T}\right) - 1}. \quad (3)$$

Here the wave vector is measured in units of reciprocal periods of the 2D lattice with the translations $\delta_{1,2}$ in Fig. 7.

The magnon spectrum $\varepsilon(\mathbf{k})$ is described in the exchange approximation by the relation (4) of Ref. [21]:

$$\varepsilon_{\mathbf{k}} = g\mu_B H + J\cos(k_x) + 2J'\cos\left(\tfrac{1}{2}k_x\right)\cos\left(\tfrac{1}{2}\sqrt{3}k_y\right) - J - 2J'. \quad (4)$$

The result of the calculation after relation (3) is shown in the upper panel of Fig. 6. This calculation is made for $\mu_0 H = 11$ T and exchange parameters J and J' for Cs_2CuCl_4 from Ref. [21]. 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 $J/(2\pi\hbar)$ was studied in the $S = \frac{1}{2}$ antiferromagnet on the distorted triangular lattice Cs_2CuCl_4. The doublet of resonance lines, marking the boundaries of the spinon continuum, was found to collapse in the field of about one-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 $S = \frac{1}{2}$ Heisenberg antiferromagnetic chain [15,24]. 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 [26]. The upper and lower boundaries of these continua for the $S = \frac{1}{2}$ Heisenberg antiferromagnetic chain in the presence of a magnetic field are calculated by Müller *et al.* These data are summarized in Table II of Ref. [15]. 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. [15], taking the boundary frequencies at $q = q_{\rm DM}$ for the boundaries with nonvanishing spectral weight. In this way we obtain the following frequency-field dependencies for spin-resonance absorption at $\mathbf{H} \parallel \mathbf{D}$:

$$2\pi\hbar f_1 = JR(h)\sin\left(\frac{q_{\rm DM}}{2}\right)\cos\left(\frac{q_{\rm DM}}{2} - \pi m(h)\right) - Jh, \quad \text{(A1)}$$

$$2\pi\hbar f_2 = JR(h)\sin\left(\frac{\pi}{2} - \frac{q_{\rm DM}}{2}\right)\cos\left(\frac{\pi}{2} - \frac{q_{\rm DM}}{2} - \pi m(h)\right), \quad \text{(A2)}$$

$$2\pi\hbar f_3 = JR(h)\sin\left(\frac{\pi}{2} + \frac{q_{\rm DM}}{2}\right)\cos\left(\frac{\pi}{2} + \frac{q_{\rm DM}}{2} - \pi m(h)\right). \quad \text{(A3)}$$

Here $h = g\mu_{\rm B}\mu_0 H/J$ is the reduced field ($h_{\rm sat} = 2$), $R(h) = \pi + h(1 - \frac{\pi}{2})$ is the field-dependent renormalization prefactor, and $m(h)$ is the reduced magnetization given by [15]

$$m(h) = \frac{1}{\pi}\arcsin\left(\frac{1}{1 - \pi/2 + \pi/h}\right). \quad \text{(A4)}$$

Domains of these functions in q and H are chosen to avoid negative values of frequencies. Using Eqs. (A1)–(A4) we get the ESR frequencies shown in Fig. 3 for the $S = \frac{1}{2}$ spin chain with uniform Dzyaloshinsky-Moriya interaction in the so-called "Müller ansatz" approximation: mode f_1 is given by the lower boundary of the S_{-+} continuum and $f_{2,3}$ are given by the lower boundary of the S_{+-} continuum. In the case of low field $h \ll 1$ these equations transform into corresponding relations of Refs. [11,13]. Modes f_2 and f_3 correspond to A′ and A in Fig. 3, and mode f_1 is relevant only for small magnetic fields which are out of range of the present study.

[1] O. A. Starykh, Rep. Prog. Phys. **78**, 052502 (2015).
[2] M. F. Collins and O. A. Petrenko, Can. J. Phys. **75**, 605 (1997).
[3] Y. Tokiwa, T. Radu, R. Coldea, H. Wilhelm, Z. Tylczynski, and F. Steglich, Phys. Rev. B **73**, 134414 (2006).
[4] O. A. Starykh, H. Katsura, and L. Balents, Phys. Rev. B **82**, 014421 (2010).
[5] R. Coldea, D. A. Tennant, A. M. Tsvelik, and Z. Tylczynski, Phys. Rev. Lett. **86**, 1335 (2001).
[6] R. Coldea, D. A. Tennant, and Z. Tylczynski, Phys. Rev. B **68**, 134424 (2003).
[7] D. Heidarian, S. Sorella, and F. Becca, Phys. Rev. B **80**, 012404 (2009).
[8] M. Kohno, O. A. Starykh, and L. Balents, Nat. Phys. **3**, 790 (2007).
[9] I. Dzyaloshinsky, J. Phys. Chem. Solids **4**, 241 (1958).
[10] T. Moriya, Phys. Rev. **120**, 91 (1960).
[11] S. Gangadharaiah, J. Sun, and O. A. Starykh, Phys. Rev. B **78**, 054436 (2008).
[12] H. Karimi and I. Affleck, Phys. Rev. B **84**, 174420 (2011).
[13] K. Yu. Povarov, A. I. Smirnov, O. A. Starykh, S. V. Petrov, and A. Ya. Shapiro, Phys. Rev. Lett. **107**, 037204 (2011).

[14] A. I. Smirnov, K. Yu. Povarov, S. V. Petrov, and A. Ya. Shapiro, Phys. Rev. B **85**, 184423 (2012).
[15] G. Müller, H. Thomas, H. Beck, and J. C. Bonner, Phys. Rev. B **24**, 1429 (1981).
[16] A. G. Gurevich and G. A. Melkov, *Magnetization Oscillations and Waves* (CRC Press, 1996).
[17] A. Abragam and B. Bleaney, *Electron Paramagnetic Resonance of Transition Ions* (Oxford University Press, 2012).
[18] A. Pimenov, A. Mukhin, V. Yu. Ivanov, V. Travkin, A. Balbashov, and A. Loidl, Nat. Phys. **2**, 97 (2006).
[19] I. A. Zaliznyak, S.-H. Lee, and S. V. Petrov, Phys. Rev. Lett. **87**, 017202 (2001).
[20] D. A. Tennant, R. A. Cowley, S. E. Nagler, and A. M. Tsvelik, Phys. Rev. B **52**, 13368 (1995).
[21] S. A. Zvyagin, D. Kamenskyi, M. Ozerov, J. Wosnitza, M. Ikeda, T. Fujita, M. Hagiwara, A. I. Smirnov, T. A. Soldatov, A. Ya. Shapiro, J. Krzystek, R. Hu, H. Ryu, C. Petrovic, and M. E. Zhitomirsky, Phys. Rev. Lett. **112**, 077206 (2014).
[22] R. Coldea, D. A. Tennant, K. Habicht, P. Smeibidl, C. Wolters, and Z. Tylczynski, Phys. Rev. Lett. **88**, 137203 (2002).
[23] M. A. Fayzullin, R. M. Eremina, M. V. Eremin, A. Dittl, N. van Well, F. Ritter, W. Assmus, J. Deisenhofer, H.-A. Krug von Nidda, and A. Loidl, Phys. Rev. B **88**, 174421 (2013).
[24] M. Kohno, Phys. Rev. Lett. **102**, 037203 (2009).
[25] M. Zhitomirsky (private communication).
[26] M. Oshikawa and I. Affleck, Phys. Rev. B **65**, 134410 (2002).