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Electron Spin Resonance of the Interacting Spinon Liquid

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We report experimental verification of the recently predicted collective modes of spinons, stabilized by backscattering interaction, in a model quantum spin chain material. We exploit the unique geometry of uniform Dzyaloshinskii–Moriya interactions in $K_2CuSO_4Br_2$ to measure the interaction-induced splitting between the two components of the electron spin resonance (ESR) response doublet. From that we directly determine the magnitude of the "marginally irrelevant" backscattering interaction between spinons for the first time.

Much of the current research in quantum magnetism is motivated by the search for an elusive quantum spin liquid (QSL) phase of the magnetic matter. A salient feature of this entangled quantum state is the presence of fractionalized elementary excitations such as fermionic spin-1/2 spinons, interactions between which are mediated by the emergent gauge field [1, 2]. This exotic, yet deeply rooted in history [3–7] perspective represents striking contrast with the usual integer-spin bosonic spin-wave excitations of the magnetically ordered media. It is firmly based on the remarkable experimental findings on (quasi) one-dimensional (1D) spin-1/2 magnetic insulators. These include observations of a particle-hole continuum of excitations (also referred to as a "two-spinon continuum") in the dynamical spin susceptibility $\chi(q,\omega)$ [8, 9] and magnetic field-controlled soft modes resulting from transitions on the Zeeman-split 1D Fermi surfaces [10, 11]. The most recent milestone of this journey is provided by the Kitaev's honeycomb model which harbors Majorana fermions as elementary excitations [12]. Experimental glimpses of this exciting physics [13–15] continue to attract intense attention from the scientific community.

Close analogy between fractionalized excitations of the QSL and those of the standard Fermi liquid contains an important caveat. Unlike the latter, elementary excitations of the QSL are highly non-local objects which appear and disappear only in pairs. Thus, the spinons can not avoid interacting with each other. Interaction between spinons, as well as the curvature of the spinon dispersion, determine shape of the continuum near its edges [16, 17]. In particular, one may expect a strong backscattering between the fermionic quasiparticles confined in 1D geometry. Still, being a "marginally irrelevant" interaction in the Renormalization Group (RG) sense [18, 19], the spinon backscattering manifests itself only through weak logarithmic corrections to the observables (e.g. the uniform spin susceptibility [20], or the nuclear magnetic relaxation rate [21]) and is barely detectable this way. As the very recent theoretical findings show, it becomes most important when magnetic field is applied, shifting spinon continuum up in energy and producing a spin-1 oscillatory collective mode of spinons [22] that originates from the Larmor frequency, a spin chain analog of the Silin spin wave in non-ferromagnetic metals [23–27]. The backscattering interaction is straightforwardly manifest here through qualitative spectrum modifications [22, 28]. However, direct observation of this novel effect with e.g. neutrons is a challenging task that requires thoroughly balancing the field strength, the magnetic energy scale of the material, and the instrument resolution. Yet, alternative spectroscopic methods can overcome these difficulties.

In this Letter we, for the first time, experimentally investigate this interaction-induced modification of the spinon continuum with the help of the electron spin resonance (ESR) technique. Our measurements lead to the direct and unambiguous determination of the backscattering interaction between fractionalized spinon excitations of the spin chain. This finding is facilitated by the unique feature of the material — the uniform Dzyaloshinskii–Moriya (DM) interaction [29, 30].

The material of our study is $K_2CuSO_4Br_2$, providing an outstanding realization of the $S = 1/2$ Heisenberg chain antiferromagnet perturbed by a small uniform DM interaction [32–35]. The magnetic Cu^{2+} spin-1/2 ions at distance $a = 7.73$ Å to each other are forming linear chains running along the **a** axis of the crystal (see inset of Fig. 1a). Antiferromagnetic interaction $J \simeq 20.5$ K [32] is mediated by a two-bromine unit, that lacks inversion center within the ac plane. This lack of inversion symmetry naturally gives rise to the small DM interaction directed along the **b** axis. Hence, it can be described as Heisenberg spin-1/2 chain, with exchange interaction J between nearest-neighbor spins, perturbed by the uniform DM interaction $\mathbf{D}\cdot\hat{\mathbf{S}}_n\times\hat{\mathbf{S}}_{n+1}$ and subject to the external magnetic field $\mathbf{H}$. We focus on the parallel geometry when the magnetic field is aligned along the DM axis (z-axis), $\mathbf{H}\parallel\mathbf{D}\parallel\hat{z}$, which preserves the symmetry of rotations about z. The Hamiltonian reads

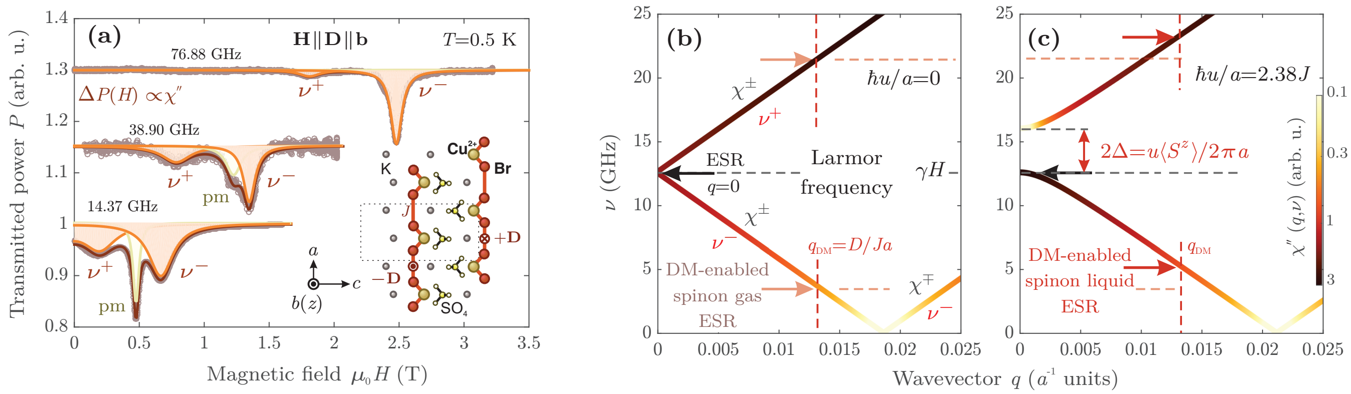

FIG. 1. Electron spin resonance in $K_2CuSO_4Br_2$. (a) Several low-T resonance lines. Magnetic field is applied along the DM axis b. Gray points is the measured rate of absorption of the microwave radiation by the sample. It is well fitted by several Lorentzian lines (dark red line). The contributions of modes ν^+ and ν^- is shown in orange color. At low frequencies an additional parasitic paramagnetic resonance can be detected (light green color). The inset shows the sketch of $K_2CuSO_4Br_2$ crystal structure. (b) Calculated spectrum of small-q transverse spin fluctuations in a magnetized spin chain without the backscattering interaction. (c) The same for the interacting spinons, $\hbar u = 2.38Ja$. For $K_2CuSO_4Br_2$ considered here $J = 20.5$ K, and the magnetic field is 0.3 T in both panels. The solid lines show $\nu(q)$ for the poles of the transverse spin susceptibility according to Ref. [22, 31], and the color shows their intensity. Black (red) horizontal arrows indicate ESR frequencies of the spin chain without (black) and with (red) the DM interaction.

$$\hat{\mathcal{H}} = \sum_n J\hat{\mathbf{S}}_n \cdot \hat{\mathbf{S}}_{n+1} - D\hat{z}\cdot \hat{\mathbf{S}}_n \times \hat{\mathbf{S}}_{n+1} - g\mu_B H \hat{S}^z_n. \quad (1)$$

Semiclassically, the competition between the antiferromagnetic Heisenberg exchange J and DM interaction $\mathbf{D}$ results in an incommensurate spiral, the period of which is determined by the wavevector $q_{\rm DM} = \tan^{-1}(D/J)/a \approx D/(Ja)$. Quantum-mechanically one can employ the unitary position-dependent rotation of spins $\hat{S}^+_n = \hat{\tilde{S}}^+_n e^{-iq_{\rm DM}na}$ which eliminates the DM term from the Hamiltonian (1) for the price of the momentum boost $q \to q + q_{\rm DM}$ [36] [31].

A *uniform*, bond-independent arrangement of DM vectors within the chain is a very rare occasion. However, a truly remarkable property of $K_2CuSO_4Br_2$ that distinguishes it from similar materials (e.g. Cs_2CuCl_4 [37, 38]) is that the DM axis is the *same*, i.e. oriented along the b crystal axis, for all spin chains [32]. This unique feature allows us to realize $\mathbf{H} \parallel \mathbf{D}$ geometry experimentally, which is a crucial element of our study. In this case the energy absorption rate measured by ESR is in fact determined by $\mathrm{Im}[\tilde{\chi}^{\pm}(q_{\rm DM}, \nu)]$ [36, 39] — the ESR becomes a finite momentum probe of the dynamic spin susceptibility!

The response of the chain (1) at small momenta can in turn be understood in terms of fermion quasiparticles - spinons [22, 40]. In the low-energy continuum limit the Heisenberg spin-1/2 chain is described by the field theory of two component Dirac spinors $\hat{\psi}_{R/L} = (\hat{\psi}_{R/L,\uparrow}, \hat{\psi}_{R/L,\downarrow})^{\rm T}$ [41]. Operators $\hat{\psi}_{R/L,s}$ describe spin-up ($s =\uparrow$) and spin-down ($s =\downarrow$) fermions with wavevectors near the right/left Fermi points $\pm k_F$ of the 1D Fermi surface. Uniform spin fluctuations are represented by the spin current operators $\hat{\mathbf{J}}_{R/L} = \frac{1}{2}\hat{\psi}^{\dagger}_{R/L}\boldsymbol{\sigma}\hat{\psi}_{R/L}$. The Hamiltonian is written as the sum of two terms, $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{V}_{\rm bs}$,

$$\hat{\mathcal{H}}_0 = \int dx \left[\hbar v_F \left(\hat{\psi}^{\dagger}_R(x)(-i\partial_x)\hat{\psi}_R(x) + \hat{\psi}^{\dagger}_L(x)(i\partial_x)\hat{\psi}_L(x)\right) - g\mu_B H\left(\hat{J}^z_R(x) + \hat{J}^z_L(x)\right)\right], \quad (2)$$

$$\hat{V}_{\rm bs} = -\hbar u \int dx\, \hat{\mathbf{J}}_R(x)\cdot\hat{\mathbf{J}}_L(x). \quad (3)$$

Here $v_F = \pi Ja/(2\hbar)$ is the spinon Fermi velocity, and u denotes the backscattering (also known as the current-current) interaction between spinons. Despite its somewhat complicated appearance $\hat{\mathcal{H}}_0$ describes a non-interacting gas of neutral fermions (spinons) $\psi_{R/L,s}$ with linear dispersion, subject to the external magnetic field. Interaction between spinons is compactly encoded in $\hat{V}_{\rm bs}$, which describes $2k_F$ scattering of right- and left-moving fermions on each other. The amplitude u of this backscattering *is the key parameter that we experimentally address in our study.*

In absence of the backscattering u interaction, the ESR spectrum of the spin chain (1) is known to be a *doublet* [36]

$$h\nu^{\pm} = |g\mu_B H \pm \frac{\pi}{2}D|, \quad (4)$$

accounting for both transverse $\chi^{\pm}$ and $\chi^{\mp}$ components of the susceptibility. Its origin can be easily understood from low-energy spectrum of the spin chain depicted in Fig. 1b. The appearance of the ESR doublet in the

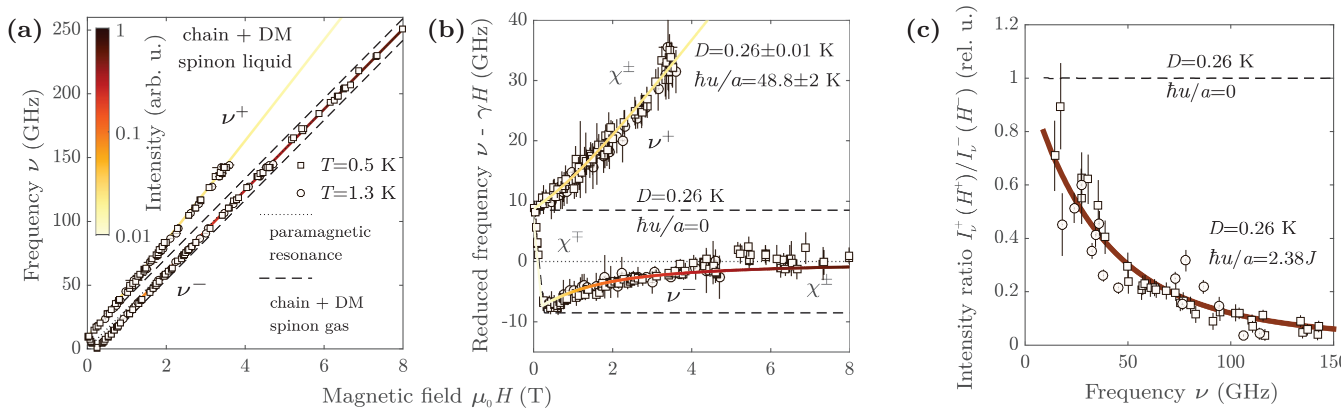

FIG. 2. ESR spectrum of $K_2CuSO_4Br_2$ and its theoretical description in terms of interacting spinons. (a) The frequency-field diagram including the $H \parallel D$ data at two different temperatures (circles and squares). Error bars are obtained from the lineshape fit and for the majority of ESR lines are estimated as 0.02 T and are within the symbol size. For some of frequencies above 50 GHz, the lineshape was distorted by the parasitic electrodynamic size effect, here errors in H-value are estimated as a whole linewidth and are seen as error bars on the left and middle panels. The dotted line is the $h\nu = g\mu_B H$ paramagnetic resonance, the dashed lines correspond to the non-interacting approximation (4) with $D = 0.26$ K. The solid lines and their color (intensity, as indicated in the inset) correspond to the interacting spinon theory (5). The best-fit value of $\hbar u/a = 48.8$ K is used. (b) The same diagram, but in the "reduced" representation with γH subtracted, $\nu \to \nu - \gamma H$. (c) The mode intensity ratio versus the observation frequency. Points are the experimental data (as in the other panels), error bars stem from the experimental uncertainty in determining the widths and amplitudes of the two overlapping peaks. Solid line is the theoretical prediction (8) for the obtained best-fit value $\delta = 0.12$. The dashed line shows non-interacting spinons result.

quantum spin liquid state of the spin chain is the fingerprint of the uniform DM interaction. It has been experimentally observed previously in three different materials, Cs_2CuCl_4 [38], the present compound $K_2CuSO_4Br_2$ [42], and also in $K_2CuSO_4Cl_2$ [43].

However, the non-interacting spinon description of the dynamic spin response is qualitatively incomplete. Similar to the case of the interacting electron liquid [23, 24], the backscattering interaction u qualitatively changes transverse spin susceptibility for small (q, ν), as demonstrated by the recent interacting spinon theory [22], supported by DMRG calculations and numerical Bethe-ansatz study [44]. The new poles of $\chi^{\pm,\mp}$ are shown in Fig. 1c. The ESR frequencies are now given by [22]

$$h\nu^\pm = \left| g\mu_B H + \Delta \pm \sqrt{\Delta^2 + (1-\delta^2)\left(\frac{\pi}{2}D\right)^2} \right|. \qquad (5)$$

Here $2\Delta = \hbar u\langle \hat{S}^z\rangle/a$ is the interaction-induced spectral gap (splitting), and $\langle \hat{S}^z\rangle/a = \chi H$ is the mean spin $z-$component per unit length. The theory is most compactly expressed in terms of the small dimensionless parameter δ,

$$\delta = \frac{1}{2a}\hbar u \frac{\chi_0}{g\mu_B} = \frac{u}{4\pi v_F}, \qquad (6)$$

that describes the enhancement of the renormalized zero-field spin susceptibility per unit length $\chi = \chi_0/(1-\delta)$, from its non-interacting value $\chi_0 = g\mu_B a/(2\pi\hbar v_F)$. Using δ, the splitting becomes:

$$\Delta = \frac{\delta}{1-\delta} g\mu_B H. \qquad (7)$$

Even when $D = 0$, Eq.(5) predicts finite spectral gap 2Δ between the $\nu^\pm$ branches, as Fig. 1c shows.

This feature does not contradict the Larmor theorem because the intensity of the upper branch ν^+ vanishes as q^2 in the $q \to 0$ limit, whereas the lower intense branch ν^-remains exactly at the Larmor frequency $h^{-1}g\mu_B H = \gamma H$. Hence, in the absence of the symmetry-breaking perturbations the Larmor theorem is actually obeyed, and backscattering interaction u makes no difference for the ESR experiment on the *ideal* spin chain. Thus, the symmetry-breaking uniform DM interaction in (1) is absolutely crucial for accessing both modes with ESR.

The experiments were done at the Kapitza Institute on a set of multifrequency (1-250 GHz) resonant cavity ESR inserts into ^{3}He and ^{4}He-pumping cryostats equipped with superconducting magnets. The transmission of microwave power P through the sample-containing resonator was measured as the function of the magnetic field at a fixed frequency ν. It is affected by the dissipative susceptibility of the spin subsystem and can be approximately expressed as $\Delta P/P \propto \chi''(0,\nu)$ [45], or rather $\chi''(q_{\rm DM},\nu)$ in the presence of the uniform DM interaction. To compare these predictions with the ESR experiments on $K_2CuSO_4Br_2$ we use two sets of data. One, taken at $T = 0.5$ K, was previously partly described in [42]. The other set, taken at at $T = 1.3$ K, was not presented before. The datasets involve multiple samples of $K_2CuSO_4Br_2$.

Several examples of the raw spectrometer microwave transmission data at 0.5 K are shown in Fig. 1a. The data demonstrates a well-resolved doublet of $\nu^{\pm}$ lines, with a parasitic line (not exceeding 25% of ν-lines intensity) in the middle that comes from impurities and is sample-dependent. The intensity of this mode shows Curie-like temperature dependence typical of impurity spins (see Ref. [42] for more details). Even on the qualitative level one can notice the increase of the distance between the ν^+ and ν^- components of the doublet with the resonance frequency, and the accompanying 'fading out' of the ν^+ mode. Both effects are in agreement with the interacting spinon picture that can be inferred from Fig. 1c and Eq. (5). For $u = 0$, the intensities of two modes are equal and the splitting is field-independent, determined by the magnitude of the DM vector only, as previously found in the low-field range below 1 T [42].

These observations can be further quantified by fitting measured ESR spectra with three overlapping Lorentzian functions to extract the precise intensities and resonant fields [31]. Results of this new data analysis presented in Fig. 2 show a striking quantitative agreement with the interacting spinon theory. The deviation between the data and the non-interacting spinon approximation (4) is clearly visible in the frequency-field diagrams of Fig. 2a,b. While being completely unexplainable within the previous non-interacting approximation (4), all the 'deviant' effects – the upward deflection and the fading of the upper ν^+ mode, and the restoration of the Larmor precession for the lower ν^- mode – are readily explained by the new interacting spinon expression (5,7). The upward shift of the ν^+ mode is the consequence of the gap Δ (7) growing with the field. The same effect is responsible for the upward approach of the ν^- mode towards the Larmor frequency, as is seen in Fig. 2.

Fitting the $\nu(H)$ data to Eq.(5), we find an excellent agreement between the experiment and the interacting-spinon theory for the value $\delta = 0.12 \pm 0.005$. By Eq.(6), this means that the backscattering interaction constant is $\hbar u/a = 48.8 \pm 2$ K. This is a strong interaction indeed, it corresponds to 2.38 ± 0.1 in the exchange coupling J units. What matters, however, is that u enters equations (5) and (7) only via δ, which is quite small. This smallness implies the consistency of the made theoretical assumptions. This is further confirmed by the spinon mean-free path estimate [46], and analysis of u in terms of RG approach [19] [31]. Note that DM interaction strength D, while being an independent parameter of the fit, is actually unchanged compared to the previous estimate 0.26 ± 0.01 K [42]. Its value is fixed by the zero field splitting $\pi D/2$ which at zero magnetization is not affected by the interaction u.

The *intensity ratio* $I^+_\nu(H^+)/I^-_\nu(H^-)$ at a fixed frequency ν (with $H^{\pm}$ being the resonant fields of the corresponding modes) is another quantity that can be determined both theoretically and experimentally. The theory [22] predicts:

$$\frac{I^+_\nu(H^+)}{I^-_\nu(H^-)} = \frac{\sqrt{(h\nu\delta)^2 + ((1-\delta^2)\pi D/2)^2} - h\nu\delta}{\sqrt{(h\nu\delta)^2 + ((1-\delta^2)\pi D/2)^2} + h\nu\delta}. \quad (8)$$

This parameter-free comparison is shown in Fig. 2c. We find an excellent agreement between all our datasets and the theory (8) for the derived value $\delta = 0.12$. Notice that without the backscattering interaction u, i.e. for $\delta = 0$, the ratio would be just 1 for all frequencies.[1]

Thus, the relative attenuation of the ν^+ mode represents an additional confirmation of the validity of the interacting spinon description of $K_2CuSO_4Br_2$.

To summarize, the observed field evolution of the ESR spectrum is very well explained by the model of interacting spinons. The normally well-hidden backscattering interaction turns out to be a crucial ingredient for both *qualitative and quantitative* description of the data. The obtained value of the spinon backscattering interaction $u \simeq 1.5v_F \simeq 3.5 \cdot 10^5$ cm/s is of the order of spinon velocity v_F. Experimental confirmation of the importance of interactions between spinons reveals a genuine Fermi-liquid-like (in contrast to a Fermi-gas-like) behavior of the quasiparticles constituting the spin chain ground state. Dynamic small-momentum response of the quantum spin chain demonstrates an amazing similarity with an electron liquid and, in particular, Silin spin waves in non-ferromagnetic conductors [23, 24]. This result paves way to spectroscopic investigations of more complex quantum spin liquids, including higher-dimensional ones [47, 48].

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[1] Note again that (8) describes the ratio of intensities of modes $\nu^{\pm}$ measured at the same fixed frequency but for different resonant fields $H^{\pm}$ while Figures 1(b,c) illustrate relative intensities of $\nu^{\pm}$ modes for the fixed magnetic field. See [31] Sec. IV.C.

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