Bound spinons in an antiferromagnetic S=\frac{1}{2} chain with a staggered field

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Inelastic neutron scattering was used to measure the magnetic field dependence of spin excitations in the antiferromagnetic S=\frac{1}{2} chain CuCl\textsubscript{2} \cdot 2(dimethylsulfoxide) (CDC) in the presence of uniform and staggered fields. Dispersive bound states emerge from a zero-field two-spinon continuum with different finite energy minima at wave numbers q=\pi and q_i \approx \pi(1 - 2(S_z)). The ratios of the field dependent excitation energies are in excellent agreement with predictions for breather and soliton solutions to the quantum sine-Gordon model, the proposed low-energy theory for S=\frac{1}{2} chains in a staggered field. The data are also consistent with the predicted soliton and n=1,2 breather polarizations and scattering cross sections.

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Shortly after the advent of quantum mechanics, Hans Bethe introduced a model antiferromagnet that continues to play a central role in quantum many body physics \cite{1}. The isotropic antiferromagnetic (AF) spin-1/2 chain, has a simple spin Hamiltonian: $H = J\sum_i S_i \cdot S_{i+1}$, is integrable through Bethe’s Ansatz, and is realized with high fidelity in a number of magnetically anisotropic Cu\textsuperscript{2+} based materials. Because it sits at the boundary between quasi-one-dimensional spin-1/2 chain systems \cite{7, 8, 9} and is now understood to be a distinguishing attribute of quantum-critical systems. The ground state is a Luttinger liquid \cite{10}, and the spinons are non-local spin-1/2 objects with short range interactions. Thus, spinons can only be excited in pairs and produce a gapless continuum. Such a spectrum confines spinons in multi-particle bound states \cite{13}. A quantitative theory for this effect was developed by Affleck and Oshikawa starting from the following extension of Bethe’s model Hamiltonian \cite{13, 14}.

$$H = J\sum_i S_i S_{i+1} + \sum_j (-1)^j D \cdot (S_{j-1} \times S_j)$$

$$- \sum_{j,\alpha,\beta} H^\alpha g^\alpha_{\alpha\beta} + (-1)^j g^s_{\alpha\beta} |S^\beta_j|^2.$$ (1)

The alternating spin environment is represented by the staggered Dzyaloshinskii-Moriya (DM) interaction and Zeeman terms. Through an alternating coordinate transformation, the model can be mapped to a spin-1/2 chain in a transverse staggered field that is proportional to the uniform field $H$. While the zero field properties of Eq.(1) are indistinguishable from Bethe’s model, an applied field induces transverse AF Ising spin order and a gap. Using bosonization techniques to represent the low energy spin degrees of freedom, Affleck and Oshikawa showed that their dynamics is governed by the quantum sine-Gordon model (QSG) with Lagrangian density

$$\mathcal{L} = \frac{1}{2} [(\partial_t \phi)^2 - (\partial_x \phi)^2] + hC \cos(\beta \phi).$$ (2)

Here $h \propto H$ is the effective staggered field. $C(H)$ and $\beta(H)$ (which goes to $\sqrt{2\pi}$ for $H \to 0$) vary smoothly with the applied field and can be determined numerically through the Bethe Ansatz for $h << H$ \cite{14, 15}

With applications from classical to particle physics, the SG model plays an important role in the theory of non-linear dynamic systems \cite{16}. Excitations are composed of topological objects called solitons that encompass a localized $\pm 2\pi/\beta$ shift in $\phi$ for a soliton and anti-soliton respectively \cite{17}. In addition there are soliton-antisoliton bound states called breathers, which drop below the soliton-anti-soliton continuum as the non-linear...
Deuterated single crystals were grown through slow cooling of saturated methanol solutions of anhydrous copper chloride and deuterated dimethyl sulfoxide ((CD$_3$)$_2$SO) in a 1:2 molar ratio. The sample studied consisted of four crystals with a total mass 7.76 g. The experiments were performed using the disk chopper time-of-flight spectrometer (DCS) at the NIST Center for Neutron Research with the c-axis and the magnetic field vertical. In configuration A the incident energy was $E_i=3.03$ meV and the a-axis was parallel to the incident beam direction $k_i$. Configuration B had $E_i=4.64$ meV and $\angle(k_i,a)=60^\circ$. The counting time was 18 hrs at 11 T and an average of 5 hrs for each measurement between 0 and 8T. The raw scattering data were corrected for a time-independent background measured at negative energy transfer, for monitor efficiency, and for the Cu$^{2+}$ magnetic form factor, folded into the first Brillouin zone, and put onto an absolute scale using the elastic incoherent scattering from CDC. For the normalization, the H/D ratio (=0.02) was measured independently through prompt-γ neutron activation analysis.

Figures 1(a) and 1(b) show that for $T \ll J/k_B$, the zero-field excitation spectrum of CDC consists of continuum scattering above a low-energy threshold that varies as $h\omega = \frac{\omega}{2}J/|\sin(q)|$ through the zone $\Gamma$. An exact analytical expression for the two spinon contribution to the scattering cross section which accounts for 72.89% of the total spectral weight was recently obtained by Bougourzi et al. In Figs. 2 and 3 show a quantitative comparison of this result (blue line), duly convoluted with the experimental resolution, to the experimental data. The excellent quantitative agreement between model and data provides compelling evidence for spinons in the zero field state of CDC. Note that the Goldstone modes that are expected due to Néel order for $h\omega < k_B T_N \approx 0.1$ meV, are not resolved in this experiment.

Figures 1(c) and 1(d) show that the magnetic excitations in CDC change dramatically with field and are dominated by resolution-limited modes for $H = 11$ T. Figures 2(a) and 3(b) show spectra at the wave vectors corresponding to the minima in the dispersion relations, which occur at $q = \pi$, and $h\omega = 0.03$ meV and the $q_i = 0.77\pi$, as determined from the constant-$h\omega$ cut in Fig. 2(a). These data graphically illustrate the field-induced transfer of spectral weight from the two-spinon continuum into single-particle excitations. A phenomenological cross-section of long-lived dispersive excitations was fit to the data near $q = \pi$ and $q_i$, to take into account the experimental resolution and thereby accurately locate the excitation energies. These fits are shown as red lines in Figs. 2 and 3. The inferred parameters characterizing the dispersion relations are displayed for a series of fields in Fig. 4.

We now examine whether our high field observations are consistent with the QSG model for spin-1/2 chains in a staggered field. First, the model predicts single soliton excitations at $q_s = \pi(1 - 2\langle S_z \rangle)$. This is qualita-
FIG. 2: Spectra at \( q = \pi \) observed with configurations A (a) and B (b) for zero field and 11 T obtained from data in the range 0.95\( \pi < q < 1.05\pi \). The zero-field intensity for 0.3\( \pi < q < 0.4\pi \) was subtracted as a background. The top axes indicates wave-vector transfer, \( k \), perpendicular to the chain. The blue line is a fit of the two-spinon cross-section to the data as explained in the text, including the theoretically calculated breather continua polarized perpendicular to the field \( H \). The inset in (b) highlights the high energy range.

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Quantitative agreement is also apparent for the unfitted peak corresponds to a moving breather. These data are from configuration B. The ranges of integration were (a) 0.675 meV \( < h\omega < 0.75 \) meV, and (b) 0.72\( \pi \approx < q < 0.82\pi \).

The soliton and antisoliton mass is related to the exchange interaction of the original spin chain and the uniform staggered field, \( H \) and \( h \), as follows [14, 17]

\[
M \approx J \frac{\hbar^2}{g_\mu_B H} (1 + \xi)/2 \times \left( B \frac{J}{g_\mu_B H} \right)^{2(\pi - \beta^2)/4\pi} (2 - \beta^2/\pi)^{1/4} - (1 + \xi)/2. \tag{3}
\]

Here \( \xi = \beta^2/(8\pi - \beta^2) \rightarrow 1/3 \) for \( H \rightarrow 0 \) and \( B = 0.422169 \). Assuming \( h \propto H \), the soliton energy versus field is shown as a solid red line in Fig. 3(a). While Eq. (3) is at the limit of validity for CDC at \( H = 11 \) T, we attribute the discrepancy with the mode energy at \( q_i \) (red triangles) to inter-chain interactions that suppress the effective staggered field close to \( H_c \). The more general expression, \( h \propto (H - H_c)\alpha \), yields a good fit for \( \alpha = 0.68(5) \) (dashed red line). Neutron scattering probes the projection of spin fluctuations on the plane normal to \( H \) and perpendicular (parallel) to the scattering vector \( \mathbf{Q} \). Figure 3 shows that the \( \hbar\omega = 1 \) meV peak seen for \( \mathbf{Q}_A \approx (1, 1.64, 0) \) in configuration A is absent for \( \hbar\omega = 1 \) meV and \( \mathbf{Q}_B \approx (1, 0, 0) \) in configuration B. In a quasi-one-dimensional system the only explanation for this is that the excitation is polarized along \( \mathbf{Q}_B |\alpha| |\mathbf{h} \) as expected for an even numbered breather, and hence is distinguished by the polarization factor in the neutron scattering cross section for configuration B. The predicted \( \mathbf{b} \) and \( \mathbf{c} \) axis polarizations respectively of the \( n=1 \) breather and the soliton are confirmed by the consistent polarization factor corrected intensities from configurations A and B for \( H = 11 \) T in Fig. 4(c).

One of the unusual aspects of the QSG model is that complex features such as the breather and soliton structure factors can be calculated exactly [15]. The solid
The peak close to 1 in configuration A and polarized along nsity predicted for the first moment continuum scattering comes from the field dependence of for experiment shows that a high energy continuum persists there. Interpretation of the data throughout the Brillouin zone will require exact diagonalization studies and a better understanding of lattice effects than provided by the continuum field theory reviewed in this paper. Other results that call for further experimental and theoretical work are the observation of high energy continuum scattering in the gapped phase and indications that inter-chain interactions can renormalize the soliton mass.

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In summary, staggered field induced spinon binding in spin-1/2 chains provides an experimental window on the unique non-linear dynamics of the quantum sine-Gordon model. Our neutron scattering experiment on quasi-one-dimensional CDC in a high magnetic field yields clear evidence for soliton/antisoliton creation at wave vector transfer \( q = \pi(1 - 2(S^2)) \), as well as \( n = 1 \) and \( n = 2 \) breather bound states at \( q = \pi \). Prediction of the theorectical result suggests that there are additional longitudinal contributions to the continuum scattering.

\[ \langle H \rangle = \frac{1}{2} \ln 2 \]

predicted by the QSG model \( 14 \) and shown as a solid line in the inset of Fig. 2 has a maximum close to a weak peak in the measured scattering intensity. The shortfall of the theoretical result suggests that there are additional longitudinal contributions to the continuum scattering.

In addition to the field-induced resonant modes, the experiment shows that these exact results are consistent with the field dependent intensities of the commensurate and incommensurate low energy modes in CDC. For \( H = 11 \) T the third breather is expected at about 1.4 meV, close to the energy of the soliton mode at \( h=1 \). The peak close to 1.4 meV has intensity \( I = 0.14(3) \) in configuration A and \( I = 0.26(2) \) in configuration B and this is consistent with a third breather contribution polarized along b. The inferred b-polarized intensity of \( I^{\text{b}} \) is however much greater than the intensity predicted for the \( n = 3 \) breather \( (I^{\text{SC}} = 0.026(7)) \), which indicates additional anisotropic contributions to the inelastic scattering there.

In addition to the field-induced resonant modes, the experiment shows that a high energy continuum persists for \( H=11 \) T. Fig. 3(d) for example shows a broad maximum in the \( q \)-dependence of neutron scattering for energies \( h\omega > 1.6 \) meV and \( q \approx \pi \). Firm evidence for continuum scattering comes from the field dependence of the first moment \( \langle h\omega \rangle = h^2 \int S(Q,\omega)\omega d\omega \), which is proportional to the ground state energy \( \langle H \rangle \) with a negative \( q \)-dependent prefactor \( 24 \). At zero field the experimental value of \( \langle h\omega \rangle \) corresponds to \( \langle H \rangle = -0.4(1)J \), in agreement with Bethe’s result of \( \langle H \rangle = (\frac{1}{4}\ln 2)J = -0.44J \).

At 11 T, however, \( \langle H \rangle \) derived solely from the resonant modes is \(-0.25(6)J \) when \( \langle H \rangle \) is expected to be \(-0.34J \) \[ 10 \]. The discrepancy is an independent indication of spectral weight beyond the resonant modes. For \( q = \pi \), the transverse contribution to the continuum scattering predicted by the QSG model \[ 14 \] and shown as a solid line in the inset of Fig. 2 has a maximum close to a weak peak in the measured scattering intensity. The shortfall of the theoretical result suggests that there are additional longitudinal contributions to the continuum scattering.

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![FIG. 4: (a) Field dependence of the soliton and breather modes. The lines are the QSG predictions for \( h \sim 0 \) (solid) and \( h=c(H-H_0)\sqrt{\hbar} \) (dashed). (b) Ratio of breather energies to soliton energies versus \( H \), compared to the QSG model. (c) Energy integrated intensities divided by the QSG-predicted polarization factors and compared to structure factor predictions using particle energies given by the dashed line in (a) and a common overall scale factor \[ 12 \]. (d) Incommensuration \( q_f \) obtained from the minimum in the soliton dispersion compared to the predicted \( q_f = \pi(1-2(S_z)) \) using the magnetization curve \( \langle S_z \rangle = 1/\pi \arcsin(1-\pi/2 + \pi J/(q_f H_0))^{-1} \) for a Heisenberg spin-1/2 chain. The field dependence was measured with configuration A, and an additional measurement at \( H = 11 \) T was performed using configuration B.](image)