Energy Separation of Single-Particle and Continuum States in an $S = 1/2$ Weakly Coupled Chains Antiferromagnet

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Inelastic neutron scattering is used to study transverse-polarized magnetic excitations in the quasi-one-dimensional $S = 1/2$ antiferromagnet $BaCu_2Si_2O_7$, where the saturation value for the Néel order parameter is $m_0 = 0.12\mu_B$ per spin. At low energies the spectrum is totally dominated by resolution-limited spin-wave-like excitations. An excitation continuum sets in above a well-defined threshold frequency. Experimental results are discussed in the context of current theories for weakly interacting quantum half-integer-spin chains.

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Studies of low-dimensional (low-D) magnetism have greatly contributed to our understanding of many-body quantum mechanics. Quantum-mechanical effects are, as a rule, enhanced in low dimensions. This is particularly true for magnetic systems, thanks to the nontrivial commutation relation of spin operators. Simple spin Hamiltonians, which can be easily studied theoretically, often demonstrate such fundamental phenomena as mass generation, hydrodynamics breakdown, topological disorder, and quantum criticality [1]. Quantum magnets are more than just toy models: a number of almost exact realizations have been found in real quasi-low-D magnetic materials, enabling experimental studies and a direct comparison with theory. Quantum magnets can also provide insight on how exotic low-D physics relates to the more familiar semiclassical picture that seems to dominate the three-dimensional (3D) world. This *dimensional crossover* can be studied, for example, in systems composed of weakly coupled spin chains. In particular, for $S = 1/2$ Heisenberg antiferromagnets (AFs), of great interest is the interplay between classical dynamics in Néel-ordered 3D materials (single-particle order-parameter excitations, or spin waves) and critical dynamics of the quantum-disordered $S = 1/2$ one-dimensional (1D) model (two-spinon excitation continuum [2,3]).

In quasi-1D $S = 1/2$ systems, for arbitrary weak interchain interactions, long-range order is restored. However, according to recent theories [4,5], as long as the 3D order parameter remains small, the excitation spectrum has a unique *dual* nature. At low frequencies the diffuse continuum is "cleaned up" and replaced by long-lived single-particle "magnon" excitations. What remains of the critical dynamics in isolated chains is now seen at higher energies as a series of multimagnon states. The lowest-energy contribution is from two-magnon processes. At the 1D AF zone center the continuum thus sets in at twice the characteristic magnon energy and is separated from the single-particle modes by an energy gap. To date, this separation of single-particle and continuum dynamics has not been clearly observed experimentally in any quasi-1D material. Among other technical obstacles is the limited choice of suitable model compounds. Most of what is experimentally known of coupled $S = 1/2$ chains comes from neutron scattering studies of $KCuF_3$ [6–9]. Here the ordered moment is rather large, making the spin waves very intense at low energies and the continuum contribution often difficult to isolate. Materials like Sr_2CuO_3 [10] or $SrCuO_2$ [11,12], on the other hand, are very good 1D systems, and it is the spin waves that are hard to identify. Below we report inelastic neutron scattering studies of spin dynamics in the $S = 1/2$ quasi-1D antiferromagnet $BaCu_2Si_2O_7$. Using neutron setups with complimentary resolution characteristics, we clearly observe the frequency separation of single-particle and continuum contributions. In doing so, we pay special attention to the polarization of magnetic excitations and compare the results to existing theoretical models.

The recently characterized $BaCu_2Si_2O_7$ (orthorhombic space group *Pnma*, $a = 6.862$ Å, $b = 13.178$ Å, $c = 6.897$ Å) appears to be an ideal model material for the present study. Compared to $KCuF_3$, it has a stronger 1D character: with an in-chain (along the c axis) Heisenberg exchange constant $J = 279$ K, the Néel temperature is only $T_N = 9.2$ K. The structure of the magnetically ordered phase was previously guessed from bulk data and an analysis of a *single* magnetic Bragg reflection [13]. As part of the present work, we performed a comprehensive neutron diffraction study of the magnetic structure at $T = 1.5$ K using the D23 single-crystal diffractometer at Institut Laue-Langevin (ILL) [14]. Overall, 35 inequivalent magnetic Bragg intensities were measured. The spin arrangement was found to be exactly as proposed in Ref. [13]. The obtained value for the ordered moment $m_0 = 0.12\mu_B$ is much smaller than in $KCuF_3$ ($m_0 \approx 0.5\mu_B$), yet large enough to expect a measurable spin-wave contribution to inelastic scattering.

Low-energy magnetic excitations in $BaCu_2Si_2O_7$ were studied in high-resolution inelastic neutron scattering experiments on a $5 \times 5 \times 50$ mm^3 $BaCu_2Si_2O_7$ single-crystal sample at the IN14 cold-neutron spectrometer at ILL [14]. Measurements were performed in the $(0, k, l)$ scattering plane with a fixed final neutron energy of 3 meV. Typical constant-Q and constant-E scans collected near the 1D AF zone center $l = 1$ are shown in Fig. 1. Figure 2 shows a series of constant-Q scans for different momentum transfers perpendicular to the chain axis. The polarization dependence of the magnetic neutron scattering cross section ensures that, to a good approximation, only fluctuations of spin components perpendicular to the ordered moment are observed. Very similar data sets (not shown) were collected using a comparable setup on the TASP cold-neutron three-axis spectrometer at Paul Scherrer Institut, Switzerland, for momentum transfers along the $(h, 0, 1)$ and $(h, h, 1)$ reciprocal-space rods [14].

The main feature seen in all constant-Q scans is a sharp asymmetric peak that we attribute to single-magnon excitations. The intensity onset on the low-energy side is very steep and suggests the peaks are resolution limited. The extended high-energy "tail" is due to wave vector resolution effects. The data were analyzed using a single-mode approximation (SMA) cross section, derived from the quantum chain–mean field (chain-MF) model [5]. The dispersion measured along $(0, k, 1)$, $(h, 0, 1)$, and $(h, h, 1)$, we found that at least three interchain exchange constants are needed, between nearest-neighbor spins along the $(1, 0, 0)$, $(0, 1, 0)$, and $(1, 1, 0)$ real-space directions, which we denote as J_x, J_y, and J_3, respectively. The SMA cross section was convoluted with the calculated experimental resolution function and used to fit all measured scans. The in-chain coupling constant J was fixed at $J = 24.1$ meV, as previously deduced from bulk susceptibility curves [13]. An excellent global fit to all scans is obtained with $J_x = -0.463(2)$ meV, $J_y = 0.161(1)$ meV, and $2J_3 = 0.145(1)$ meV. Simulations based on these values are shown in solid lines in Figs. 1 and 2. The resulting dispersion relation along the $(0, k, 1)$ direction is shown as a solid line in the $(\hbar\omega)$-k plane in Fig. 2. Additional analysis also gives an upper estimate for the intrinsic excitation width: 0.03 meV. Up to about 4 meV energy transfer the magnetic excitation spectrum is thus fully accounted for by the single-particle picture.

Extending the cold-neutron study to higher energy transfers was hindered by certain geometrical instrumental constraints. Instead, the intermediate-energy spectrum was studied using the IN22 thermal-neutron spectrometer at ILL using 14.7 meV final-energy neutrons. Most of the data were collected in constant-E scans along the chain axis, in the vicinity of the $(0, 0, 3)$ 1D AF zone center. The background originating from the empty Al sample holder was measured separately for each scan. Typical background-subtracted data sets are presented in Fig. 3. None of these show the distinct two-peak structure as at 3 meV energy transfer (Fig. 1c). To verify that this is not a resolution effect, we performed a least-squares fit of the SMA cross section, convoluted with the calculated resolution, to each measured profile. Very poor fits are obtained, as shown by the dashed lines in Fig. 3. It is clear that if the observed intensity was due to a single mode, two separate peaks would have been easily resolved. For lack of a convenient analytical result for interacting chains, we fitted the measured scans to the Müller-ansatz continuum

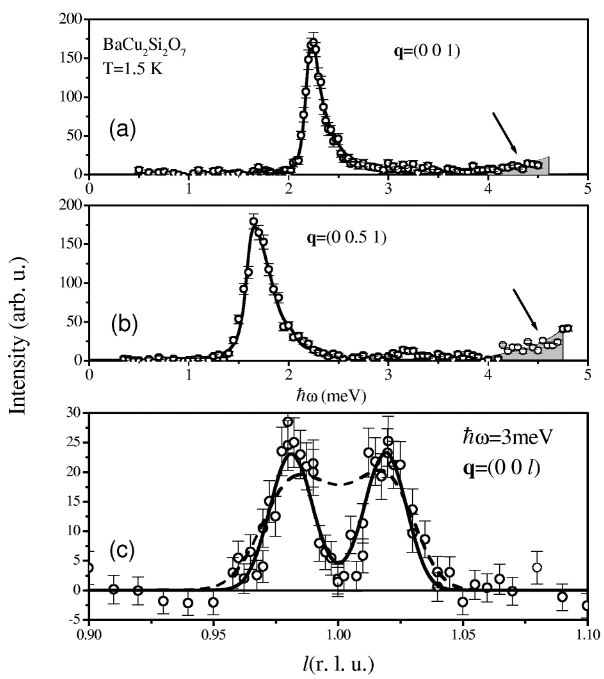

FIG. 1. Typical constant-Q (a),(b) and constant-E (c) scans measured in $BaCu_2Si_2O_7$ at $T = 1.5$ K near the 1D AF zone center $l = 1$ (symbols). Lines and shaded areas indicated by arrows are as described in the text.

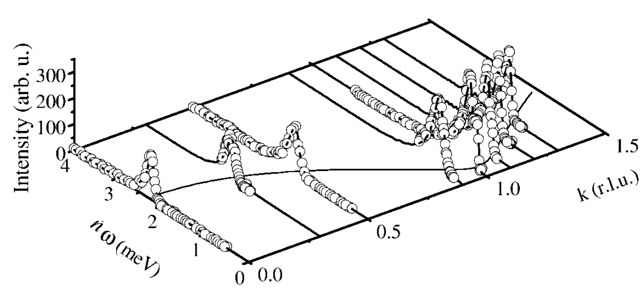

FIG. 2. A series of constant-Q scans measured in $BaCu_2Si_2O_7$ at $T = 1.5$ K for different momentum transfers perpendicular to the chain axis. Lines are as described in the text.

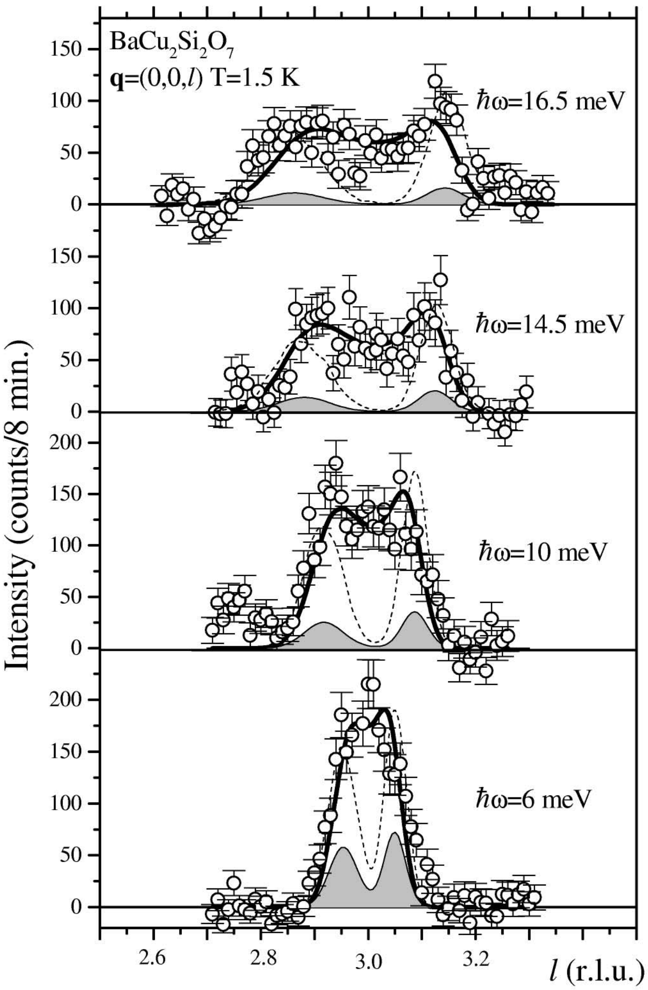

FIG. 3. Constant-E scans measured in $BaCu_2Si_2O_7$ at $T = 1.5$ K using a thermal-neutron setup. Lines are fits to the data, as discussed in the text. Shaded areas are the extrapolated single-mode contributions.

form [15], known to work well for isolated chains. The agreement in this case is much better (solid lines in Fig. 3). For contrast, the dashed line in Fig. 1c shows the best Müller-ansatz fit to the 3 meV high-resolution constant-E scan. Unlike at low energies, a large contribution to the observed scattering at high energies must thus come from continuum excitations.

Additional evidence was obtained in analyzing the measured intensities. In the chain-MF model the intensity of magnon scattering has the same $1/\omega$ dependence as for classical antiferromagnetic spin waves, in full agreement with the bulk of our cold-neutron data. We exploited this scaling relation to estimate spin-wave intensities at higher energy transfers. A normalization factor was obtained by fitting the SMA cross section to the intensity measured using the thermal setup at $\boldsymbol{q} = (0, 0, 3)$ between 2 and 3 meV transfer, where, according to our cold-neutron results, the SMA picture is still valid. The thus calculated single-mode contributions are represented by shaded areas in Fig. 3. Clearly, magnon excitations account for only a minor fraction of the dynamic susceptibility at high frequencies. In Fig. 4a we plot the measured q_z-integrated intensity I_z, scaled by energy transfer $\hbar\omega$, as a function of the latter. The advantage of this integration procedure that the actual wave-vector dependence of continuum scattering becomes unimportant. The expected spin-wave contribution is practically constant above 3 meV in this plot (Fig. 4a, dashed line). This follows from that in a fixed-final energy configuration one directly measures $S(\boldsymbol{q}, \omega)$ without any additional energy-dependent scaling factors ($\lambda/2$ effects in the monitor are estimated to be less than 5%). In contrast, experimentally, $I_z\omega$ increases substantially with frequency.

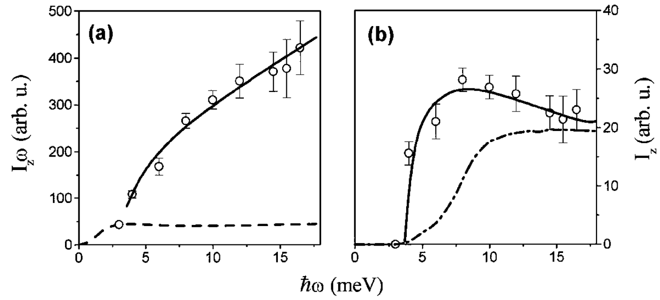

FIG. 4. (a) Measured q_z-integrated intensity scaled by energy transfer (symbols). The dashed line is the extrapolated single-mode contribution. (b) Estimated q_z-integrated intensity of continuum excitations in $BaCu_2Si_2O_7$ (symbols). The dash-dotted line is the calculated spin-wave theoretical three-magnon spectrum. In both panels the solid line is a guide for the eye.

The continuum fraction can be isolated by subtracting the extrapolated SMA part from the experimental data. The result is shown in Fig. 4b. The energy resolution being about 1.5 meV, we conclude that the continuum sets in between 3.5 and 5.5 meV energy transfer. We have verified that this result is quite robust and remains valid even if we assume that there is a 50% systematic error in the estimate of the single-mode contribution. From Fig. 4b we see that above the threshold the q_z-integrated continuum intensity is only slightly energy independent. Looking back at the cold-neutron data, we find that they actually contain direct evidence of continuum scattering. Indeed, the increase of intensity seen around 4.5 meV in Fig. 1 is *not* accounted for by the SMA picture and likely represents the onset of the continuum.

To understand the observed spectra, we recall the simple yet profound physical picture provided by the quantum chain-MF theory [4,5]. For an isolated chain in a staggered field, the magnons have an energy gap Δ that scales as $H_\pi^{2/3}$ [4,16,17]. In the coupled-chain case, magnon dispersion perpendicular to the chain axis leads to a softening of the gap at the 3D zone center for transverse excitations. These gapless modes correspond to conventional spin waves in the classical spin-wave theory (SWT) and are the long-lived excitations seen in $BaCu_2Si_2O_7$ at low energies. Excitations at exactly Δ can be observed in points of reciprocal space where interchain interactions cancel out at the MF level. In $BaCu_2Si_2O_7$ this occurs at

$\boldsymbol{q} = (0.5, 0.5, 1)$. Using the measured exchange constants we can estimate $\Delta = 2.37$ meV, in perfect agreement with a direct measurement [14]. It is straightforward to verify that the experimental values for Δ, J, m_0, and T_N agree with chain-MF predictions [4] remarkably well.

A unique quantum-mechanical feature of a half-integer Heisenberg AF chain in a staggered field, totally absent in conventional SWT, is a *longitudinal* mode, i.e., a magnon excitation polarized parallel to the direction of static staggered magnetization. The existence of such an excitation was recently confirmed experimentally in $KCuF_3$ [9]. Focusing on transverse spin correlations in the present study, we do not directly observe this branch in $BaCu_2Si_2O_7$. It does, nonetheless, play an important role in the observed transverse *continuum*. In the chain-MF model for weakly interacting chains, low-energy continuum excitations are seen as multimagnon states [5]. The existence of a longitudinal mode allows *two*-particle excitations in the transverse channel, composed of one longitudinal and one transverse magnon. The threshold for such excitations at the 1D AF zone center is at twice the characteristic magnon energy: $\Delta_c = 2\Delta$. This prediction is consistent with our experimental results for $BaCu_2Si_2O_7$, where Δ_c is between 3.5 and 5.5 meV, and $2\Delta = 4.8$ meV.

An alternative model that predicts multimagnon continua is the conventional nonlinear SWT. In $BaCu_2Si_2O_7$, however, the ordered moment is small, and the ground state deviates significantly from the Néel state, the starting point of any SWT calculations. In this case SWT loses self-consistency. For example, the calculated spin-wave correction to sublattice magnetization in $BaCu_2Si_2O_7$ exceeds 100% and destroys the long-range order altogether. SWT also gives an inadequate description of *transverse*-polarized excitations. The longitudinal mode being absent in this model, two-magnon transverse excitations are prohibited by selection rules. The lowest-energy contribution is from three-magnon states. The continuum should thus set in at around 3Δ. The three-magnon SWT contribution can be exactly evaluated. Following the recipe given in Ref. [18], the spin-wave bandwidths measured in $BaCu_2Si_2O_7$ were used to calculate the three-magnon profile at $\boldsymbol{q} = (0, 0, 3)$. The result (arbitrary scaling) is shown in Fig. 4b in a dash-dotted line. The calculated continuum onset is clearly at a larger energy, by 2 to 3 meV, than observed experimentally.

In summary, we find that the transverse excitation spectrum in $BaCu_2Si_2O_7$ is divided into two well-defined regions. Below 4 meV the weight is entirely consolidated into long-lived single-particle excitations. These excitations carry only a small fraction of the total spectral weight at higher energies, where a continuum of states becomes the dominant contribution. While further studies of the lower continuum bound are needed, the present results suggest a threshold between 3.5 and 5.5 meV. For this strongly 1D material the conventional SWT description becomes inadequate. In contrast, the chain-MF model, based on quantum-mechanical properties of individual chains, is in very good agreement with experiment.

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