Energy separation of single-particle and continuum states in a $S = 1/2$ weakly-coupled chains antiferromagnet.

A. Zheludev(1), M. Kenzelmann(2), S. Raymond(3), E. Ressouche(3), T. Masuda(4), K. Kakurai(5), S. Maslov(1), I. Tsukada(4,6), K. Uchinokura(4) and A. Wildes(7),

(1) Physics Department, Brookhaven National Laboratory, Upton, NY 11973-5000, USA. (2) Oxford Physics, Clarendon Laboratory, Oxford OX1 3PU, UK. (3) DRFMC/SPSMS/MDN, CENG, 17 rue des Martyrs, 38054 Grenoble Cedex, France. (4) Department of Applied Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan. (5) Neutron Scattering Laboratory, Institute for Solid State Physics, The University of Tokyo, Ibaraki 319-1106, Japan. (6) Present address: Central Research Institute of Electric Power Industry, 2-11-1, Iwato kita, Komae-shi, Tokyo 201-8511, Japan. (7) Institut Laue-Langevin, Ave. des Martyrs, Grenoble Cedex 9, France.

(March 21, 2022)

Inelastic neutron scattering is used to study transverse-polarized magnetic excitations in the quasi-one-dimensional $S = 1/2$ antiferromagnet BaCu$_2$Si$_2$O$_7$, 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.

75.40.Gb, 75.50.Ee, 75.10.Jm

One of the outstanding problems in quantum magnetism today is that of dimensional crossover in systems composed of weakly-coupled spin chains. In particular, for $S = 1/2$ Heisenberg antiferromagnets (AFs), the interplay between classical dynamics in Néel-ordered three-dimensional materials (single-particle order-parameter excitations, or spin waves) and critical dynamics of the quantum-disordered $S = 1/2$ one-dimensional (1D) model (2-spinon excitation continuum [1]) is not fully understood. In quasi-1D $S = 1/2$ systems, for arbitrary weak inter-chain interactions, long-range order is restored. However, according to recent quantum-mechanical calculations [2], 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 multi-magnon states. The lowest-energy contribution is from two-magnon processes. At the 1D AF zone-center the continuum is thus expected to set in at twice the characteristic magnon energy, and should be separated from the single-particle part of the spectrum by a “second 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$_3$ [3]. Here the ordered moment is rather large, making the spin waves very intense at low energies, and the continuum contribution difficult (though not impossible) to isolate. Materials like Sr$_2$CuO$_3$ [4] or SrCuO$_2$ [5,6], 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$_2$Si$_2$O$_7$. 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$_2$Si$_2$O$_7$ (orthorombic space group Pnma, $a = 6.862 \, \text{Å}$, $b = 13.178 \, \text{Å}$, $c = 6.897 \, \text{Å}$) 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 $J = 279 \, \text{K}$, the Néel temperature is only $T_N = 9.2 \, \text{K}$. The structure of the magnetically ordered phase was previously guessed from bulk data and an analysis of a single magnetic Bragg reflection [7]. As part of the present study, we performed a comprehensive neutron diffraction study of the magnetic structure at $T = 1.5 \, \text{K}$ using the D23 single-crystal diffractometer at Institut Laue-Langevin (ILL) [8]. Overall, 35 inequivalent magnetic Bragg intensities were measured. The spin arrangement was found to be exactly as proposed in Ref. [2]. The obtained value for the ordered moment $m_0 = 0.12 \mu_B$ is much smaller then 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$_2$Si$_2$O$_7$ were studied in high-resolution inelastic neutron scattering experiments on a $5 \times 5 \times 50 \, \text{mm}^3$ BaCu$_2$Si$_2$O$_7$ single-crystal sample at the IN14 cold-neutron spectrometer at ILL [9]. 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 mo-
momentum transfers perpendicular to the chain-axis. The polarization dependence of the magnetic neutron scattering cross section ensures that within this range, to a good approximation, only fluctuations of spin components perpendicular to the chains (and thus the ordered moment, which is parallel to c) are observed. Very similar data sets (not shown) were collected using a comparable setup on the TASP cold-neutron 3-axis spectrometer at Paul Scherrer Institut, Switzerland, for momentum transfers along the (h, 0, 1) and (h, h, 1) reciprocal-space rods. 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 a steep dispersion along the chain axis and a non-zero wave vector resolution in that direction. The data were analyzed using a single-mode approximation (SMA) cross section, derived from the quantum chain-Mean Field (chain-MF) model. The dispersion measured along (0, k, 1), (h, 0, 1) and (h, h, 1), we found that at least three inter-chain exchange constants are needed, between nearest-neighbor spins along the (1, 0, 0), (0, 1, 0) and (1, 1, 0) real-space directions, that 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 to determine $J_x$, $J_y$ and $J_3$. The in-chain coupling constant $J$ was fixed at $J = 24.1$ meV, as previously deduced from bulk susceptibility curves. 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. 1b, 1c. The resulting dispersion relation along the $(0, k, 1)$ direction is shown in a solid line in the $(h\omega)$-k plane in Fig. 2. Additional analysis also gives an upper estimate for the intrinsic excitation width: 0.03 meV. These results conclusively demonstrate that up to about 4 meV energy transfer the magnetic excitation spectrum is fully accounted for by the single-particle picture, in the entire range of momentum transfers perpendicular to the chain-axis.

![FIG. 1](image1.png)

**FIG. 1.** Typical constant-Q (a,b) and constant-E (c) scans measured in BaCu$_2$Si$_2$O$_7$ at $T = 1.5$ K near the 1D AF zone-center $l = 1$ (symbols). Lines are as described in the text.

Extending the cold-neutron study to higher energy transfers was hindered by certain geometrical constrains imposed by the design of IN14 spectrometer. 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 2-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 in 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 form, 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. 3 shows the best Müller-ansatz fit to the 3 meV high-resolution const-E scan. From this comparative analysis we conclude that, unlike at low energies, a large contribution to the observed scattering at high energies must come from continuum excitations.

Additional evidence was obtained in analyzing the measured intensities. In the chain-MF model the in-
tensity 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. Having confirmed the single-mode picture at low energies, 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 $q = (0, 0, 3)$ 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.

![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.](image)

Clearly, magnon excitations account for only a minor fraction of the dynamic susceptibility at high frequencies. To quantify this behavior, 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 \(S(\mathbf{q}, \omega)\) without any additional energy-dependent scaling factors ($\lambda / 2$ effects in the monitor are expected to be less than 5%, thanks to the use of a thermal guide). In contrast, experimentally, \(I_z \omega\) increases substantially with frequency.

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 measurements, we find that they actually do contain direct evidence of continuum scattering as well. Indeed, the increase of intensity seen around 4.5 meV in Fig. 1 is not accounted for by the SMA picture, and thus represents the onset of the continuum, in full agreement with the thermal-neutron results.

![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-dot line is the calculated spin wave theoretical 3-magnon spectrum. In both panels the solid line is a guide for the eye.](image)

To understand the observed long-lived and continuum excitations, we shall recall the simple yet profound physical picture provided by the quantum chain-MF theory. For an isolated chain in a staggered field, the magnons have an energy gap $\Delta$ that scales as $H^{2/3}_\pi$. In the coupled-chains case, magnon dispersion perpendicular to the chain axis leads to a softening of the gap at the 3D zone-center for transverse-polarized excitations. These gapless modes correspond to conventional spin waves in the classical spin wave theory (SWT), and are the long-lived excitations that we see in BaCu_2Si_2O_7 at low energies. Excitations at exactly $\Delta$ can be observed in points of reciprocal space where inter-chain-
interactions cancel out at the MF level. In BaCu$_2$Si$_2$O$_7$ this occurs at $q = (0.5, 0.5, 1)$. Using the measured exchange constants we can estimate $\Delta = 2.37$ meV. It is straightforward to verify that the experimental values for $\Delta$, $J$, $m_0$ and $T_N$ satisfy the relations predicted by the chain-MF model \[8\] 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$_3$ \[13\]. Focusing on transverse spin correlations in the present study, we do not directly observe this branch in BaCu$_2$Si$_2$O$_7$. It does, nonetheless, play an important role in the observed transverse continuum. In the chain-MF and field-theoretical models for weakly-interacting chains, low-energy continuum excitations are seen as multi-magnon states \[14\]. 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$_2$Si$_2$O$_7$, where $\Delta_c$ is between 3.5 and 5.5 meV, and $2\Delta = 4.8$ meV.

An alternative model that predicts multi-magnon continua is SWT with kinematic corrections, which explicitly takes into account non-linearities in the spin wave Hamiltonian. Continuum excitations observed in KCuF$_3$ are remarkably well described by this model \[16\]. In BaCu$_2$Si$_2$O$_7$, however, the ordered moment is much smaller, and the ground state resembles the Néel phase, i.e., the starting point of SWT calculations, considerably less. For this material SWT completely fails at the quantitative level. For example, the spin wave correction to sublattice magnetization in BaCu$_2$Si$_2$O$_7$ that we calculated using the known dispersion bandwidths is well over 100%. For the transverse-polarized excitation continuum, the inadequacy of SWT becomes even more apparent. Indeed, 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 lower bound of the continuum should thus be at roughly $3\Delta$. The 3-magnon SWT contribution can be exactly evaluated, provided the spin wave dispersion is known. Following the recipe given in Ref. \[15\], the spin wave bandwidths measured in BaCu$_2$Si$_2$O$_7$ were used to calculate the 3-magnon profile at $q = (0, 0, 3)$. The result (arbitrary scaling) is shown in Fig. 3 in a dash-dot 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$_2$Si$_2$O$_7$ is divided into two well-defined regions. Below 4 meV the weight is consolidated into long-lived single-particle spin wave-like excitations, and no sign of multi-particle continuum is found. These excitations carry only a small fraction of the total spectral weight at higher energies, where an excitation continuum 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 spin wave-theoretical description becomes inadequate, even if kinematic corrections are taken into account. In contrast, the chain-MF model, based on quantum-mechanical properties of individual chains, is in very good agreement with experiment.

We would like to thank Dr. P. Böni (PSI Villigen) for allowing us to refer to the yet unpublished TASP data, Dr. L.-P. Regnault (CEA Grenoble) for his assistance with experiments at ILL, Prof. A. Tsvelik, Prof. R. A. Cowley(Oxford University) and Dr. I. Zaliznyak (BNL) for illuminating discussions, and Mr. R. Rothe (BNL) for technical support. This work is supported in part by the U.S.-Japan Cooperative Program on Neutron Scattering, Grant-in-Aid for COE Research “SCP coupled system” from the Ministry of Education, Science, Sports, and Culture. Work at Brookhaven National Laboratory was carried out under Contract No. DE-AC02-98CH10886, Division of Material Science, U.S. Department of Energy. One of the authors (M. K.) is supported by a TMR-fellowship from the Swiss National Science Foundation under contract no. 83EU-053223.

\begin{thebibliography}{15}
\bibitem{1} L. D. Fadeev and L. A. Takhtajan, Phys. Lett. \textbf{85 A}, 375 (1981).
\bibitem{2} F. D. M. Haldane and M. R. Zirnbauer, Phys. Rev. Lett. \textbf{71}, 4055 (1993).
\bibitem{3} H. J. Schulz, Phys. Rev. Lett. \textbf{77}, 2790 (1996).
\bibitem{4} F. H. L. Essler, A. M. Tsvelik, and G. Delfino, Phys. Rev. B \textbf{56}, 11001 (1997).
\bibitem{5} K. Satija \textit{et al.}, Phys. Rev. B \textbf{21}, 2001 (1980).
\bibitem{6} S. E. Nagler \textit{et al.}, Phys. Rev. B \textbf{44}, 12361 (1991); D. A. Tennant \textit{et al.}, Phys. Rev. B \textbf{50}, 4003 (1993).
\bibitem{7} D. A. Tennant \textit{et al.}, Phys. Rev. B \textbf{52}, 1381 (1995).
\bibitem{8} B. Lake, D. A. Tennant and S. E. Nagler, cond-mat/9910459.
\bibitem{9} K. M. Kojima \textit{et al.}, Phys. Rev. Lett. \textbf{78}, 1787 (1997).
\bibitem{10} M. Matsuda \textit{et al.}, Phys. Rev. B \textbf{55}, R11953 (1997).
\bibitem{11} I. A. Zaliznyak \textit{et al.}, Phys. Rev. Lett. \textbf{83}, 5370 (1999).
\bibitem{12} T. Tsukada \textit{et al.}, Phys. Rev. B \textbf{60}, 6601 (1999).
\bibitem{13} Details of this and other experiments referred to here, will be reported elsewhere.
\bibitem{14} G. Müller, H. Thomas, M. W. Puga, and H. Beck, J. Phys. C: Solid State Phys. \textbf{14}, 3399 (1981).
\bibitem{15} M. Oshikawa and I. Affleck, Phys. Rev. Lett. \textbf{79}, 2883 (1997).
\end{thebibliography}
[16] D. C. Dender et al., Phys. Rev. Lett. 79, 1750 (1997).
[17] J.-i. Igarashi and A. Watabe, Phys. Rev. B 43 13456 (1991); corrected in J.-i. Igarashi, Phys. Rev B 46, 10763 (1992).