

## Dominance of the Excitation Continuum in the Longitudinal Spectrum of Weakly Coupled Heisenberg $S = 1/2$ Chains

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A low-field spin-flop transition in the quasi-one-dimensional antiferromagnet  $\text{BaCu}_2\text{Si}_2\text{O}_7$  is exploited to study the polarization dependence of low-energy magnetic excitations. The measured longitudinal spectrum is best described as a single broad continuum, with no sharp “longitudinal mode,” in apparent contradiction with the commonly used chain-mean-field and random phase approximation (MF/RPA) theories. The observed behavior is also quite different than that previously seen in the related  $\text{KCuF}_3$  material, presumably due to a large difference in the relative strength of interchain interactions. The results highlight the limitations of the chain-MF/RPA approach.

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The physics of weakly interacting spin chains is of great fundamental interest, as it represents the crossover from quantum to semiclassical spin dynamics. Excitations in conventional three-dimensional magnets are single-particle spin wave states. These can be described as precessions of magnetic moments around their equilibrium orientation, and are necessarily polarized perpendicular to the order parameter. In contrast, the excitation spectrum of one-dimensional (1D) quantum-disordered antiferromagnets (AFs) is isotropic. In particular, in the  $S = 1/2$  Heisenberg model, the spectrum is a polarization-independent multispinon continuum [1–5]. Arbitrary weak interchain interactions in *quasi-1D*  $S = 1/2$  AFs restore long-range order (LRO) [6–9], break rotational invariance, and reinstate the transverse spin waves at low frequencies [10,11]. The continuum persists in the high-frequency regime.

A counterintuitive novel feature of weakly interacting Heisenberg  $S = 1/2$  chains is the *longitudinal mode* (LM), a long-lived “spin wave” polarized *parallel* to the direction of ordered moment [10,11]. This new type of excitation was recently discovered in the  $\text{KCuF}_3$  compound [12,13], where it is seen as a single symmetric broad peak in the longitudinal spectrum. [12] Such behavior is not fully understood within the commonly used chain-mean-field [6] and random phase approximation (MF/RPA) theories [10,11] that predict a sharp (long-lived) LM and a separate broad longitudinal continuum (LC). To further probe the limitations of the MF/RPA approach, and learn more about the exotic LM, we performed a polarization-sensitive inelastic neutron scattering study of  $\text{BaCu}_2\text{Si}_2\text{O}_7$ . The advantage of this material is a considerably smaller ratio of interchain and in-chain interactions, as compared to  $\text{KCuF}_3$ . This brings us closer to the idealized weak-coupling limit, where the MF/RPA theory can be expected to work best. Surprisingly, the measured longitudinal spectrum substantially deviates

from theoretical predictions. Contrary to expectation, the LM in  $\text{BaCu}_2\text{Si}_2\text{O}_7$  is altogether ill-defined, and the low-energy longitudinal spectrum is best described as a single broad continuum feature. Thus, for longitudinal polarization, there is no analogue of the separation of single-particle and continuum states, which was previously seen in the transverse spectrum [14,15].

The magnetism of  $\text{BaCu}_2\text{Si}_2\text{O}_7$  is by now very well characterized using bulk methods [16,17], neutron diffraction [16,18], and inelastic neutron scattering [14–16,19]. The material is orthorhombic (space group  $Pnma$ ,  $a = 6.862 \text{ \AA}$ ,  $b = 13.178 \text{ \AA}$ ,  $c = 6.897 \text{ \AA}$ ) with slightly zigzag AF  $S = 1/2$  chains of  $\text{Cu}^{2+}$  ions running along the  $c$  axis. The in-chain exchange constant is  $J = 24.1 \text{ meV}$ . Interactions between the chains are much weaker, and the characteristic bandwidth of spin wave dispersion perpendicular to the chain direction (“mass gap” in the terminology of Ref. [11]) is  $\Delta = 2.51 \text{ meV}$ .  $\text{BaCu}_2\text{Si}_2\text{O}_7$  orders antiferromagnetically at  $T_N = 9.2 \text{ K} = 0.033J/k_B$  with a zero- $T$  saturation moment of only  $m_0 = 0.15\mu_B$ . For comparison, for  $\text{KCuF}_3$ ,  $J = 37 \text{ meV}$ ,  $T_N = 39 \text{ K} = 0.09J/k_B$ ,  $m_0 = 0.54\mu_B$  (Refs. [3,4,20] and references therein). Because of a weak easy-axis anisotropy in  $\text{BaCu}_2\text{Si}_2\text{O}_7$ , the ordered staggered magnetization is aligned along the  $c$  axis.

The most direct way to determine the polarization of magnetic excitations is by using polarized neutrons. Such measurements are typically associated with severe intensity losses in the polarizing monochromator and analyzer. Given the size of currently available single-crystal samples, and the expected scattering cross section for longitudinal excitations in  $\text{BaCu}_2\text{Si}_2\text{O}_7$ , this type of experiment does not appear feasible. Fortunately though, even for an unpolarized neutron beam, the scattering intensity is polarization dependent. Only spin components perpendicular to momentum transfer contribute to the cross section. One commonly used strategy is to

measure the inelastic signal at equivalent wave vectors in different Brillouin zones (BZs). Comparing such data sets can provide polarization information, but relies heavily on the exact knowledge of the spectrometer resolution function. The latter can be estimated quite accurately, but an error of only a few percent can ruin the polarization analysis. This was the main problem in previous inconclusive attempts to apply this technique to longitudinal excitations in  $\text{BaCu}_2\text{Si}_2\text{O}_7$ , as described in Ref. [15].

In the present work, the problem was overcome by using the “reverse” approach. The *same* inelastic scan (always in the same BZ) was performed for *different* orientations of the ordered moment in the system. Polarization information was then extracted in a point-by-point data analysis that is robust and insensitive to any resolution effects. As described in detail below, to change the direction of ordered moment we exploited one of the recently discovered field-induced spin-re orientation transitions in  $\text{BaCu}_2\text{Si}_2\text{O}_7$  [18]. A 5 mm diameter and 40 mm long cylindrical single-crystal sample was mounted on the thermal three-axis spectrometer PONTA (5G) at the JRR3-M reactor at Japan Atomic Energy Research Institute at Tokai. The sample cylinder axis is close to the (1,1,0) reciprocal-space direction and was oriented vertically in the experiment. The (0,0,1) and (-0.5, 1.86, 0) reciprocal-lattice vectors determined the horizontal scattering plane. The sample environment was a horizontal-field 6 T cryomagnet. The field was at all times applied along the  $c$  axis of the crystal. The data were collected in an energy scan close to the 1D AF zone center (0,0,1). The design of the horizontal-field magnet imposes stringent constraints on experimental geometry, allowing only narrow  $\pm 15^\circ$  windows for the incident and scattered beams. For this reason, each data set was collected while simultaneously scanning the energy transfer  $\hbar\omega$  and momentum transfer perpendicular to the chain axis  $\mathbf{q}_\perp$ , at a constant momentum transfer along the chains  $q_\parallel = 2\pi/c$ , which is the 1D AF zone center. Figure 1 shows the trajectory of the scan relative to the previously determined spin wave dispersion in  $\text{BaCu}_2\text{Si}_2\text{O}_7$ . Also shown is the evolution of the instrument resolution ellipsoid, calculated in the Cooper-Nathans approximation [21,22].

The scan was repeated in three independent measurements. The first measurement (Fig. 2, solid circles) was performed in a magnetic field  $H_1 = 1.2$  T at  $T = 2$  K. Under these conditions, just as at  $H = 0$ , the ordered moment in  $\text{BaCu}_2\text{Si}_2\text{O}_7$  is parallel to  $c$  [18,19]. The measured intensity is related to the longitudinal (parallel to the ordered moment) and transverse (perpendicular to the ordered moment) dynamic structure factors  $S^\parallel(\mathbf{q}, \omega)$  and  $S^\perp(\mathbf{q}, \omega)$  through  $I_1(\mathbf{q}, \omega) \propto S^\perp(\mathbf{q}, \omega)(1 + \cos^2\alpha) + S^\parallel(\mathbf{q}, \omega)\sin^2\alpha + \mathcal{B}(\mathbf{q}, \omega)$ . Here  $\alpha$  is the angle between the momentum transfer  $\mathbf{q}$  and the chain axis, and  $\mathcal{B}(\mathbf{q}, \omega)$  is the background, assumed to be of nonmagnetic origin. In our particular case  $\alpha \lesssim 15^\circ$  through the entire scan, and

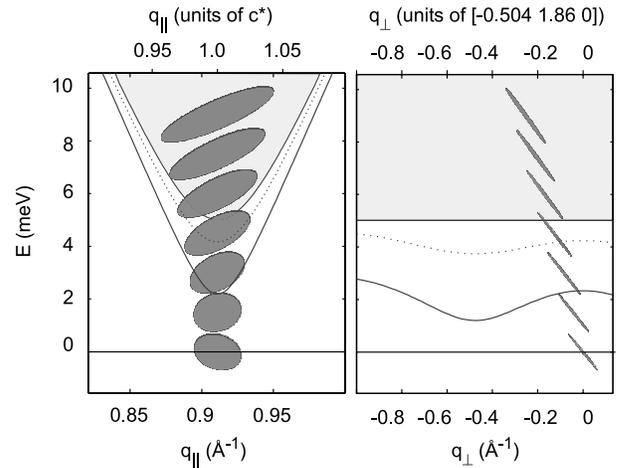


FIG. 1. ( $E - \mathbf{q}$ )-space trajectory of the inelastic scan and the corresponding evolution of the FWHM resolution ellipsoid (shaded ellipses). The solid curve shows the dispersion of spin waves in  $\text{BaCu}_2\text{Si}_2\text{O}_7$ . The dotted line and light shaded area represent the dispersion of the longitudinal mode and the domain of the excitation continuum in  $\text{BaCu}_2\text{Si}_2\text{O}_7$ , respectively, as predicted by the MF/RPA model.

it is mostly the transverse spin fluctuations that are observed. In the second measurement, the same scan was repeated at  $H_2 = 2.2$  T, which exceeds the critical field  $H_c = 2.0$  T of a spin-flop transition [17,18]. These data are plotted with open symbols in Fig. 2. In the spin-flop state, the ordered staggered moment is perpendicular to the field direction and therefore lies within the crystallographic ( $a, b$ ) plane. The measured inelastic intensity can be written as  $I_2(\mathbf{q}, \omega) \propto S^\perp(\mathbf{q}, \omega)(1 + \sin^2\alpha) + S^\parallel(\mathbf{q}, \omega)\cos^2\alpha + \mathcal{B}(\mathbf{q}, \omega)$ . For small  $\alpha$ , one observes an

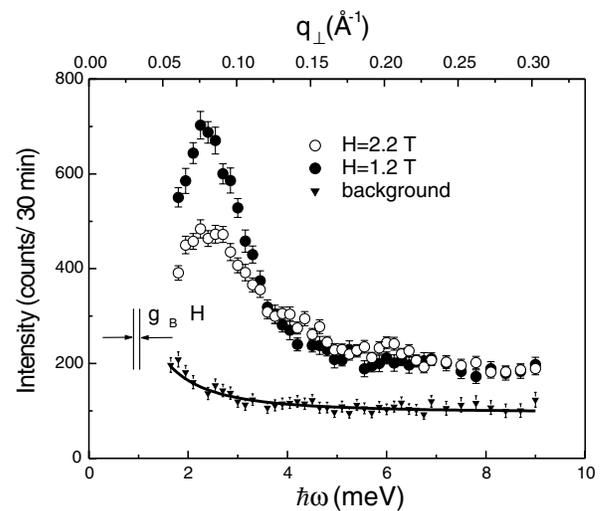


FIG. 2. Inelastic scans measured in  $\text{BaCu}_2\text{Si}_2\text{O}_7$  near the 1D AF zone center (0, 0, 1) at  $T = 2$  K in magnetic fields applied along the  $c$  axis. The data were collected below (solid circles) and above (open circles) the spin-flop transition at  $H_c = 2.0$  T. The background (triangles) was measured as described in the text. The solid line is a Lorentzian fit to the background scan.

equal mixture of longitudinal and transverse spin fluctuations. In the final measurement, performed at zero field, the sample was removed from the magnet and sample holder. The same scan was repeated to directly measure the extrinsic background  $\mathcal{B}$ , mainly due to spurious scattering in the sample environment (triangles in Fig. 2). The intrinsic background (from scattering in the sample itself) was previously shown to be negligible [15]. At each  $(\mathbf{q}, \omega)$  point, the equations for  $I_1$  and  $I_2$  were then solved to extract the dynamic structure factors for different polarizations, as plotted in Fig. 3.

Using this method to determine the polarization of spin fluctuations is based on several assumptions. The first is that the spin structure rotates *as a whole* when going through the spin-flop transition, but is otherwise unaffected. For  $\text{BaCu}_2\text{Si}_2\text{O}_7$ , such behavior was confirmed by previous neutron diffraction studies in magnetic fields up to 5 T [18]. In particular, upon crossing  $H_c$ , the magnitude of the ordered moment remains almost constant, and the change in relative alignment of nearby spins is insignificant. The second requirement is that magnetic anisotropy

and Zeeman effects are negligible on the experimentally relevant energy scales, as defined by the gap  $\Delta$  and the experimental energy resolution (typically 1.7 meV FWHM). In our measurements, this condition is also satisfied. Indeed, at  $H = H_2 = 2.2$  T the Zeeman energy per spin is an order of magnitude smaller:  $gS\mu_B H \sim 0.25$  meV. Performing differential measurements at  $H_1$  and  $H_2$ , with  $gS\mu_B \delta H$  of only 0.13 meV, further minimizes the effect, as shown by arrows in Fig. 2. The magnitude of magnetic anisotropy in  $\text{BaCu}_2\text{Si}_2\text{O}_7$  is also negligible, the associated spin wave gap being only about 0.2 meV [15]. In summary, the spin-flop transition changes only the coordinate system in which the terms “longitudinal” and “transverse” are defined, but should not affect the nature of the excitation spectrum.

Previous measurements of *transverse* spin fluctuations in  $\text{BaCu}_2\text{Si}_2\text{O}_7$  [14,15] were found to be in remarkably good quantitative agreement with MF/RPA theory [10,11]. In this approach, each individual spin chain is viewed as subject to an effective staggered exchange field  $\mathbf{H}_\pi$ , generated by LRO neighboring chains. The staggered field, in turn, generates an attractive potential between spinons, producing two-spinon bound states. Two components of the spin-triplet bound state are polarized perpendicular to  $\mathbf{H}_\pi$ . These are analogues of conventional spin waves, and have a gap  $\Delta \propto H_\pi^{2/3}$ . The third component is the longitudinal mode with a gap  $\Delta_\parallel = \sqrt{3}\Delta$ . Regardless of polarization, a multimagnon continuum, the remains of the spinon continuum, has a gap of  $\Delta_c = 2\Delta$ . Starting from this MF solution, the RPA is used to calculate the spin wave dispersion perpendicular to the chains. For  $\text{BaCu}_2\text{Si}_2\text{O}_7$ , the MF/RPA dynamic structure factor for transverse spin waves is explicitly written out in Eqs. 4–7 of Ref. [15]. The continuum part is well approximated by the “truncated Muller ansatz” functional form, as given by Eqs. 10 and 11 in the same reference. As an important consistency check, this model cross section function, after a numerical convolution with the spectrometer resolution, was fit to the transverse spectrum measured in this work. Since all parameters of the model were previously determined with very good accuracy [15,19], the only parameter varied in the present data analysis was an overall intensity scaling factor. An excellent fit was obtained, and is shown in a solid line in Fig. 3(a). Continuum and single-mode parts are shown as hatched and greyed areas, respectively.

We now turn to discussing the main result of this work, namely, the measured longitudinal spectrum. As shown in Fig. 3(b), the scattering is a broad asymmetric peak with a maximum around 4 meV energy transfer, and an extended “tail” on the high-energy side. As a first step, we analyzed the data using a model cross section based on MF/RPA theory, as for transverse excitations. The corresponding dynamic structure factor for the LM is given by Eqs. 12 and 13 of Ref. [15]. At the particular wave vector, the MF/RPA predicts the LM at 4.2 meV, quite close to the

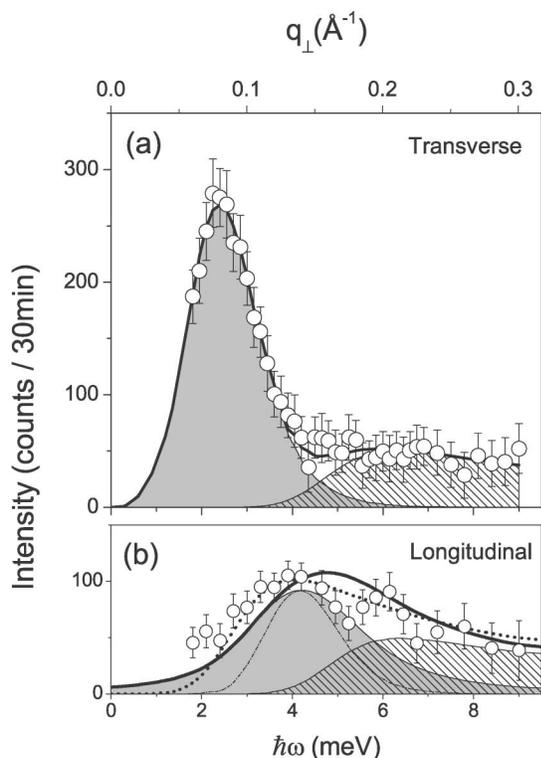


FIG. 3. Transverse-polarized (a) and longitudinal-polarized (b) dynamic structure factor measured in  $\text{BaCu}_2\text{Si}_2\text{O}_7$  near the 1D AF zone center. The heavy solid line is a fit based on MF/RPA theory, as described in the text. The gray and hatched areas represent the single-mode and continuum parts of the cross section. In (b), the heavy dotted line is a fit that does not include any single-mode contribution, as described in the text. The thin dash-dotted line is the experimental energy resolution.

observed intensity maximum. Given the shape of the observed peak, in our fits the LM was allowed to have a nonzero intrinsic energy width. The corresponding  $\delta$  function in Eq. 12 of Ref. [15] was replaced with a Lorentzian profile of FWHM  $\Gamma$ . In addition, the prefactor  $\gamma$  that characterizes the energy-scaled intensity of the longitudinal mode relative to that of transverse spin waves was treated as an adjustable parameter. Continuum scattering was assumed to be exactly as for transverse polarization. This model, when convoluted with the spectrometer resolution function, yields a reasonably good two-parameter fit to the data with  $\gamma = 2.2(1)$  and  $\Gamma = 2.4(7)$  meV [heavy solid line in Fig. 3(b)].

The obtained parameter values clearly indicate the failure of the MF/RPA model to describe longitudinal excitations in  $\text{BaCu}_2\text{Si}_2\text{O}_7$ . First, the refined intensity coefficient  $\gamma$  is about 4.5 times larger than the MF/RPA prediction [11]. Second, to obtain a good fit, we had to assume a substantial intrinsic width for the LM. In contrast, the LM in the MF/RPA model is infinitely sharp. For *weakly* coupled chains such as those in  $\text{BaCu}_2\text{Si}_2\text{O}_7$ , this damping effect has a particularly important consequence: The LM becomes altogether ill-defined. Indeed, in this case, the continuum is relatively strong, and comparable in intensity to single-mode scattering. In the MF/RPA, the LC has a (pseudo)gap at  $2\Delta$ , and the LM is centered at about  $1.73\Delta$ . At the same time, our analysis shows that in  $\text{BaCu}_2\text{Si}_2\text{O}_7$  the intrinsic FWHM of the LM is itself about  $\Delta$ . Given these numbers, the LM and the LC *are too close to be separated*, and constitute a *single continuum feature*. To illustrate this fact, in Fig. 3(b) we also show a fit to the longitudinal data using *only* the truncated Muller ansatz (heavy dotted line), with no single-mode contribution. With a continuum gap  $\Delta_c = 1.4(1)\Delta$  (smaller than the expected MF/RPA value of  $2\Delta$ ), this empirical function describes the data at least as well as the cross section that also includes a damped LM. The situation is quite different for materials with interchain interactions of intermediate strength, such as  $\text{KCuF}_3$ . Here the LC, which is the high-energy tail seen in constant- $q$  scans, is relatively weak. As a result, even though the LM is broadened, it is seen as a well-defined symmetric inelastic peak[12].

Comparing the spectra measured in  $\text{BaCu}_2\text{Si}_2\text{O}_7$  and  $\text{KCuF}_3$ , we conclude that the MF/RPA picture based on distinct LM and LC contributions does *not* become more accurate in the weak-coupling limit. The width of the LM peak normalized by  $\Delta$  in the weaker-coupled  $\text{BaCu}_2\text{Si}_2\text{O}_7$  system is even greater than in the  $\text{KCuF}_3$  compound. This counterintuitive behavior may be related to the critical nature of spin correlations in isolated  $S = 1/2$  chains. Quantum criticality implies that there is no characteristic energy scale in the chains, and therefore *any* interchain coupling is to be considered as *strong*. Strictly speaking, the RPA is well justified only for interchain interactions small compared to the energy gap in

individual chains. In our case, the chains are intrinsically gapless. The gap  $\Delta$  emerges only in the MF theory and is necessarily of the same order of magnitude as interchain coupling itself [10]. The problem is therefore only marginally suited for the RPA for intrinsic reasons.

In summary, despite a small ratio of interchain and in-chain interactions, the low-energy longitudinal spectrum of  $\text{BaCu}_2\text{Si}_2\text{O}_7$  is dominated by a broad excitation continuum, and there appears to be no distinct “longitudinal mode.” Further theoretical calculations beyond the MF/RPA are needed to better understand this behavior.

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