Short-range correlations in quantum frustrated spin system

A. Sytcheva,[1] O. Chiatti,[1,*] J. Wosnitza,[1] S. Zherlitsyn,[1] A. A. Zvyagin,[2,3] R. Coldea,[4] and Z. Tylczynski[5]
[1]*Hochfeld-Magnetlabor Dresden, Forschungszentrum Dresden–Rossendorf, D-01314 Dresden, Germany*
[2]*Max-Planck-Institut für Physik komplexer Systeme, D-01187 Dresden, Germany*
[3]*B.I. Verkin Institute for Low Temperature Physics and Engineering, Lenin Avenue 47, Kharkov 61103, Ukraine*
[4]*Oxford Physics, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, United Kingdom*
[5]*Institute of Physics, Adam Mickiewicz University, Umultowska 85, 61614 Poznan, Poland*
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We report on results of sound-velocity and sound-attenuation measurements in the low-dimensional spin-1/2 antiferromagnet Cs_2CuCl_4 (T_N=0.6 K), in external magnetic fields up to 15 T, applied along the b axis, and at temperatures down to 300 mK. The experimental data are analyzed with a theory based on exchange-striction coupling resulting in a qualitative agreement between theoretical results and experimental data.

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During the last decades low-dimensional spin systems came into the focus of solid-state physics. The main reason for this is the possibility to compare the results of experimental investigations with nonperturbative theoretical approaches available for low-dimensional spin systems. Experimental research activity was also stimulated by the technological progress in the synthesis of new materials with low-dimensional electronic properties.

It is well known that in typical three-dimensional ferromagnets (FM) and antiferromagnets (AFM) low-energy excitations are magnons (or spin waves). Magnons are quantized inhomogeneous small deviations from the classical steady-state configurations, which are associated with the quantum ground state of three-dimensional magnetically ordered systems. Each magnon is related to the reduction in the total spin of the system by 1, hence, it is accepted that magnons carry spin 1. For FM the classical steady-state configuration coincides with the exact quantum ground state because the ground state of a ferromagnet corresponds to the highest value of total spin. In contrast, for AFM the quantum ground state is related to the state with the lowest value of total spin (in most cases it is a singlet state). It is very challenging to determine the ground state for a multispin interacting system, and, therefore, it is known only for few models. For instance, it is exactly known for the Heisenberg antiferromagnetic spin-1/2 chain.[1] For this quantum system the low-energy excitations are not magnons but spinons.[2] Unlike magnons, spinons carry spin 1/2, i.e., "fractional spins." Notice, however, that spinons, due to topological reasons, can be created only in pairs. Two spinons can form a singlet state (with spin zero) or a triplet state (with spin 1). The properties of a triplet state of spinons resemble those of magnons. The uncertainty in the determination of the ground state for spin systems with antiferromagnetic interactions resulted in some theories, which use other than the standard Néel-type pictures for the lowest-energy state of AFM. Many of those models, which bring into play interacting spinons, are valid for one-dimensional spin chains. Such theories describe a quantum state known as a spin liquid. The concept of this new quantum phase was developed by different authors[3–6] after the work of Anderson,[7] where he postulated a quantum-disordered resonating-valence-bond ground state in spin-1/2 Heisenberg triangular AFM. A spin liquid is expected to exist especially in quasi-low-dimensional magnets, where quantum and classical fluctuations, enhanced due to peculiarities in the densities of states, destroy magnetic ordering at any nonzero temperatures. Also the deviations from the classical Néel description should be mostly pronounced in magnetic systems with spin frustration. Here, spin-spin interactions compete with each other producing a large degeneracy of eigenstates. In this way the structure of the ground-state and low-energy excitations are different from the Néel state.

Cs_2CuCl_4 has attracted a lot of interest as a candidate for showing spin-liquid behavior in the triangular $S=1/2$ AFM. The observation of a strong inelastic continuum in neutron scattering[8] gave rise to the vivid discussions about the nature of the ground state in Cs_2CuCl_4. It admits different spin-liquid-based interpretations,[3,4,9] as well as plausible explanations in the framework of nonlinear spin-wave theory.[10,11] Although a quantum spin liquid is strictly defined only in the ground state, its effect can be seen at finite temperatures and was suggested in case of Cs_2CuCl_4 slightly above the Néel temperature, $T_N(B)$.[8,12]

Cs_2CuCl_4 has an orthorhombic ($Pnma$) crystal structure. The planar triangular-lattice layers of Cu^{2+} spins are arranged on top of each other with a large interplanar distance along the a direction (Fig. 1). The in-plane bc interactions $J=4.34$ K along b and $J'=0.34J$ along the zigzag bonds are dominant with respect to the intraplanar interaction $J''=0.045J$. These values were estimated from the measured magnon dispersion in the saturated FM phase.[8]

Specific-heat,[13,14] susceptibility, and magnetization[14] measurements provided detailed information on the magnetic field versus temperature phase diagram. The long-range AFM order appears below $T_N \approx 0.6$ K and is coplanar with spins forming incommensurate spirals along the b direction. In applied magnetic fields several ordered phases have been observed.[14] For field along the b axis above $B_s(T=0\ \mathrm{K}) \approx 8.9$ T the system enters the fully polarized phase. In the intermediate temperature range $T_N(B) < T \lesssim 2.8$ K the compound demonstrates spin-liquid hallmarks. At about 2.8 K, the magnetic susceptibility shows a broad maximum and above this temperature it obeys the Curie-Weiss law.[14]

In this work, we present results on ultrasound investigations of Cs_2CuCl_4 performed at low temperatures down to

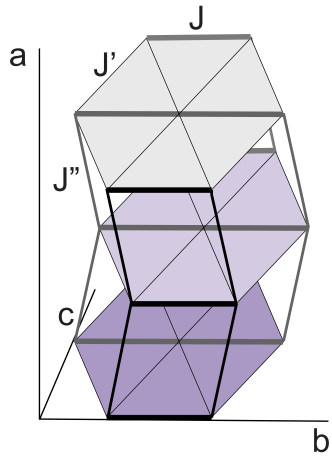

FIG. 1. (Color online) Sketch of the magnetic interactions between the Cu^{2+} ions in the Cs_2CuCl_4 structure. The Cu triangles are parallel to the *bc* plane. The largest interaction *J* is along the *b* axis.

300 mK and in magnetic fields up to 15 T. Above 100 K, the temperature dependence of the elastic constants for this compound was reported by Nasyrov *et al.*[15]

Measurements of ultrasound properties give useful information about various phase transitions and critical phenomena. See Ref. 16 for a review. For our ultrasound experiment we used a small solution-growth single crystal of Cs_2CuCl_4 from the same batch of samples as used in Ref. 13. Piezoelectric film transducers have been glued to the surfaces parallel to the *ac* crystallographic plane[17] of the sample. This geometry corresponds to the longitudinal c_{22} acoustic mode, with the wave vector **k** and polarization **u** parallel to the crystallographic *b* axis. The size of the sample along the *b* direction was 1.9 mm. The magnetic field was applied along the *b* axis, i.e., along the direction of sound propagation.

We have measured the relative change in the sound velocity and sound attenuation. The absolute value of the sound velocity at liquid helium temperature has been determined as $v_l = (2880 \pm 20)$ m/s.

The temperature dependences of the acoustic characteristics below 5 K, measured at different magnetic fields, are shown in Fig. 2. For magnetic fields lower than about 6 T, the sound velocity increases toward lower temperatures. The slope of this velocity change becomes smaller in increasing field until above 6 T the acoustic mode shows a softening and develops anomalies around the Néel temperature. These anomalies disappear above about 9 T. The sound attenuation increases steeply toward the lowest temperatures for curves taken between 6 T and B_s.

In Fig. 3, we present the magnetic field dependences of the acoustic characteristics, measured at different temperatures. The common feature of the curves is that the sound velocity exhibits a softening of the lattice stiffness with increasing field. Below about 1.5 K anomalies develop in the vicinity of the saturation field. In the fully polarized state the sound velocity almost does not change with increasing field. The sound attenuation shows a pronounced peak at about 8 T. All observed anomalies become smoother with increasing temperature. We should admit that already at 300 mK the acoustic characteristics are smeared.

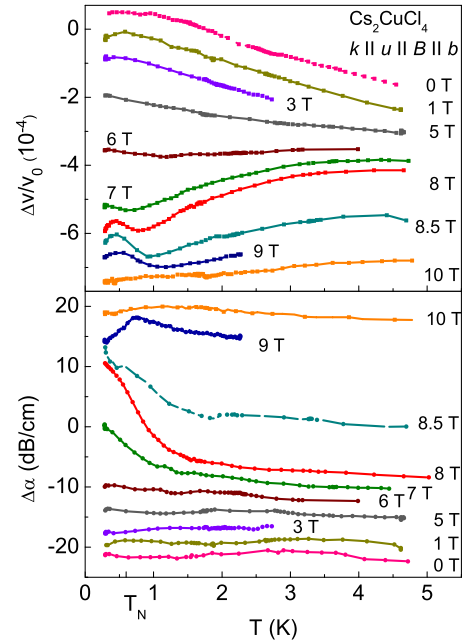

FIG. 2. (Color online) Temperature dependence of the relative change in the sound velocity and sound attenuation in different magnetic fields probed with a longitudinal ultrasound signal at a frequency of 42 MHz. The experimental geometry: $B \| k \| u \| b$. The curves are arbitrary shifted for the sake of better visibility.

The following theoretical calculation has been used to explain the observed experimental data. In magnetic materials the dominant contribution to the spin-lattice interactions mostly arises from the exchange-striction coupling. From in-

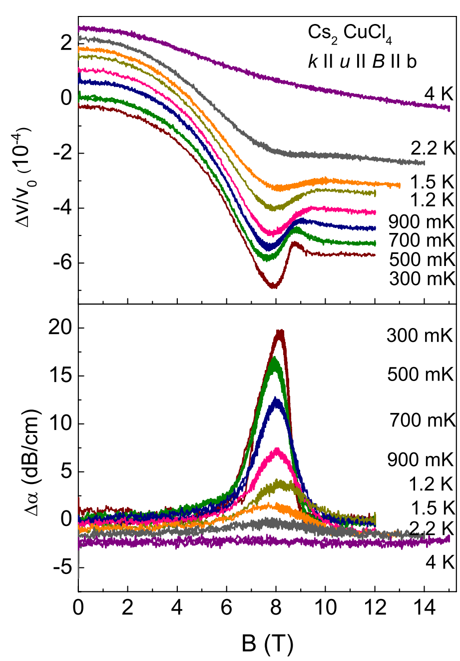

FIG. 3. (Color online) Field dependence of the relative change in the sound velocity and sound attenuation at different temperatures measured with an ultrasound signal at a frequency of 42 MHz. The experimental geometry: $B \| k \| u \| b$. The curves are arbitrary shifted for the sake of better visibility.

tuitive considerations one can expect essential changes in the magnetoacoustic characteristics of Cs_2CuCl_4 along the b axis because the longitudinal-acoustic c_{22} wave modulates the largest exchange interaction J. In our calculations we assume that in Cs_2CuCl_4 the spatial dependence of the magnetic anisotropy (i.e., of the magnetic relativistic interaction) is weaker than the spatial dependence of the exchange integrals. In this case, one can expect that mostly longitudinal sound waves interact with the spin subsystem. We approximated our system as a spin-1/2 model, in which (isotropic in spin subspace) exchange integrals between nearest-neighbor spins have different values along different directions of the crystal (spatial anisotropy). The magnetoacoustic interaction is considered then in the standard way in the framework of the perturbation approach.[18] According to Refs. 18 and 19, the renormalization of the longitudinal sound velocity of such a model can be written as

$$\frac{\Delta v}{v} = -\frac{(A_1 + A_2)}{(N\omega_{\mathbf{k}})^2}, \tag{1}$$

where

$$A_1 = 2|G_0^z(\mathbf{k})|^2 \langle S_0^z \rangle^2 \chi_0^z + T \sum_{\mathbf{q}} \sum_{\alpha=x,y,z} |G_{\mathbf{q}}^{\alpha}(\mathbf{k})|^2 (\chi_{\mathbf{q}}^{\alpha})^2,$$

$$A_2 = H_0^z(\mathbf{k}) \langle S_0^z \rangle^2 + \frac{T}{2} \sum_{\mathbf{q}} \sum_{\alpha=x,y,z} H_{\mathbf{q}}^{\alpha}(\mathbf{k}) \chi_{\mathbf{q}}^{\alpha}. \tag{2}$$

Here, N is the number of spins in the system, $\mathbf{q}$ is the wave vector of magnetic excitations, $\omega_{\mathbf{k}} = vk$ is the low-k dispersion relation with sound velocity v in the absence of spin-phonon interactions, $\langle S_0^z \rangle$ is the average magnetization along the direction of the magnetic field, $\chi_{\mathbf{q}}^{x,y,z}$ are nonuniform magnetic susceptibilities, and the subscript 0 corresponds to $q=0$. For spin systems with antiferromagnetic interactions, such as Cs_2CuCl_4, the main contribution to the summation over $\mathbf{q}$ in Eq. (2) should come from terms with $q^{x,y,z} = \pi$.

The magnetic characteristics of the spin model, which are present in Eq. (2), were calculated in two (known for Cs_2CuCl_4) ways. In the first one, we supposed quasi-two-dimensional spin model with the weak interaction between spins, belonging to different planes (considered in the random-phase approximation, as usually, cf. Ref. 19). The in-plane two-dimensional spin-spin interaction in this case we supposed to be spatially isotropic. We considered that interaction in the framework of the quasi-two-dimensional hard-core boson theory for low-energy spin excitations of the spin subsystem of Cs_2CuCl_4, like the one, developed in Ref. 13. Notice that hard-core bosons reveal the collective behavior similar to fermions (because no two fermions, as well as no two hard-core bosons, can be situated at the same site of the lattice; this property is connected with the fact that for each spin $\frac{1}{2}$ only one spin "turn" is possible). This is why, the behavior of the magnetoacoustic characteristics of the considered model reminiscent in some sense to the one, calculated in Ref. 19, where the free fermionic approximation for spin excitations was used. On the other hand, the difference between the present approach, and the one of Ref. 19 is clear. In the former case the features at the critical magnetic fields were related to the van Hove square-root singularities at the edges of the excitation band of the one-dimensional chain, while in the present case the feature (also related to the van Hove singularity) is logarithmic, as usual for the two-dimensional model. The results are shown in Fig. 4 (left panel). In Fig. 4 (right panel) we present results of another model calculation, were we took in standard way[1] spin-1/2 antiferromagnetic chains (with spinons as elementary excitations) as the first approximation for Cs_2CuCl_4 (following Ref. 9 or also Ref. 20). Interchain interactions were taken into account perturbatively. Notice that for spin-1/2 antiferromagnetic chains elementary spin excitations (spinons) also can be considered as (interacting) hard-core bosons even on better ground than in the isotropic two-dimensional case, cf. Refs. 1, 9, and 20. We use our theories only for temperatures beyond the ordering phase, naturally. Thus, for $T > T_N(B)$ these theories imply the presence of two-dimensional (quasi-one-dimensional) spin-spin correlations, i.e., at least in the latter case, the characteristic of a spin liquid. Notice that the spin-liquid behavior is expected, according to some approaches, in the two-dimensional spin systems, too, see the discussion above.

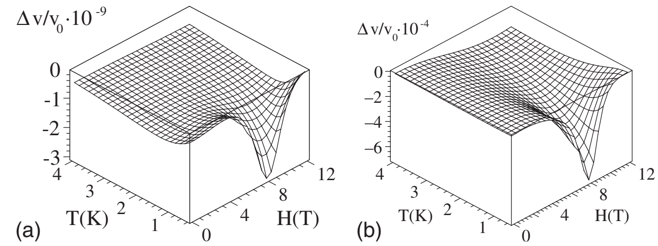

FIG. 4. Calculated field and temperature dependences of the relative change in the sound velocity for the quasi-two-dimensional isotropic (left panel) and anisotropic (right panel) spin systems.

The renormalization parameter $A_1 + A_2$ is proportional to the spin-phonon coupling constants (which have to be determined independently)

$$G_{\mathbf{q}}^{\alpha} = \frac{1}{m} \sum_n e^{i\mathbf{q}\mathbf{R}_{nm}} (e^{i\mathbf{k}\mathbf{R}_{nm}} - 1) \mathbf{e}_{\mathbf{k}} \frac{\partial J_{mn}^{\alpha}}{\partial \mathbf{R}_m},$$

$$H_{\mathbf{q}}^{\alpha} = \frac{1}{m} \sum_n e^{-i\mathbf{q}\mathbf{R}_{nm}} (e^{i\mathbf{k}\mathbf{R}_{nm}} - 1)(e^{-i\mathbf{k}\mathbf{R}_{nm}} - 1) \times \mathbf{e}_{\mathbf{k}} \mathbf{e}_{-k} \frac{\partial^2 J_{mn}^{\alpha}}{\partial \mathbf{R}_n \, \partial \mathbf{R}_m}. \tag{3}$$

Here, m is the mass of the magnetic ion, J_{mn}^{α} denote exchange integrals, $\mathbf{e}_{\mathbf{k}}$ is the polarization of the phonon with wave vector $\mathbf{k}$, and $\mathbf{R}_n$ is the position vector of the nth site.[18,19] In our calculations we used these quantities as fitting parameters. Our simplified theories reproduce the main features of the experimentally observed behavior. Namely, both model calculations manifest the pronounced minimum of the magnetic field dependence of $\Delta v/v_0$ at B_s, the shift of the position of the minimum in $\Delta v/v_0(B)$ to larger field values for higher temperatures, and the decrease in the relative sound velocity with growing field at high temperatures. With increasing T, the features near the critical field become

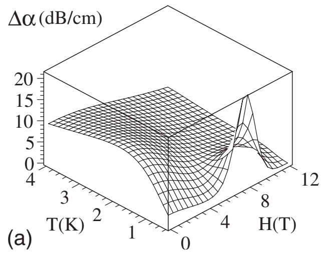

FIG. 5. Calculated field and temperature dependences of the sound attenuation for the quasi-two-dimensional isotropic (left panel) and anisotropic (right panel) spin systems.

weaker, the same way as it is observed in the experiment (cf. Fig. 3). At the phase boundary $T_N(B)$ the susceptibility of the system diverges, and our theories predict very narrow and high peaks at the critical value of B_s (not shown). Therefore, for the sake of clarity, the curves in Fig. 4 are plotted only for $T > T_N(B)$. Notice, however, the better agreement between the results of calculations and the experimentally observed behavior in the case, where the spin-spin in-plane interactions were spatially anisotropic (quasi-one-dimensional situation).

Following Refs. 18 and 19, we also calculated the attenuation coefficient, which reads as

$$\Delta\alpha(\equiv \Delta\alpha_k) = \frac{1}{Nv}\left[2|G_0^z(\mathbf{k})|^2 \langle S_0^z\rangle^2 \chi_0^z \frac{\gamma_0^z}{(\gamma_0^z)^2 + \omega_{\mathbf{k}}^2} + T\sum_{\mathbf{q}} \sum_{\alpha=x,y,z} |G_{\mathbf{q}}^\alpha(\mathbf{k})|^2 (\chi_{\mathbf{q}}^\alpha)^2 \frac{2\gamma_{\mathbf{q}}^\alpha}{(2\gamma_{\mathbf{q}}^\alpha)^2 + \omega_{\mathbf{k}}^2} \right], \qquad (4)$$

where $\gamma_{\mathbf{q}}^\alpha$ are the relaxation rates, which were approximated as $\gamma_{\mathbf{q}}^\alpha = \beta / T\chi_{\mathbf{q}}^\alpha$, where β is a material-dependent constant (see Ref. 18). In our calculations we supposed that the relaxation rates do not depend on the direction and on the wave vector. The results are presented in Fig. 5 for the spatially isotropic quasi-two-dimensional spin model (left panel) and for the spatially anisotropic, quasi-one-dimensional in-plane spin-spin interactions (right panel). Again, our theories reproduce the main features, observed in the experiment: an abrupt increase in the sound attenuation near the saturation field B_s, which becomes less pronounced at higher temperatures. All these results demonstrate the important role magnetic excitations play in the vicinity of $T_N(B)$, revealing the short-range magnetic order in the studied range of temperatures. Again, notice that the quasi-one-dimensional approximation (where the spin-liquid state seems natural) reproduces the features of the magnetic and temperature behavior of the sound attenuation better than the calculations, based on spatially isotropic in-plane spin-spin interactions. Our calculations are valid beyond the long-range ordering region of the phase diagram but at low temperatures below around 2.6 K. This shows the onset of the short-range spin-spin correlations in Cs_2CuCl_4

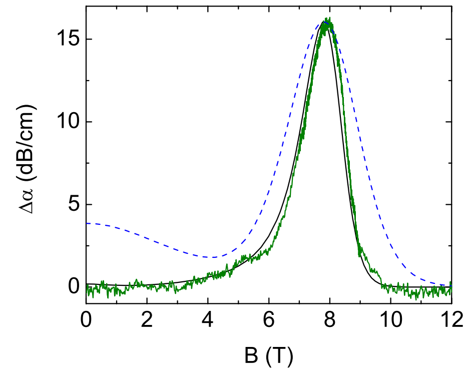

FIG. 6. (Color online) Magnetic field dependence of the sound attenuation, calculated in the framework of the model with spatially anisotropic in-plane spin-spin interactions (black solid line), spatially isotropic interactions (blue dashed line), and experimentally observed data (green solid line) for $T = 500$ mK.

for $T > T_N(B)$. On the other hand, in the ferromagnetic part of the phase diagram, the agreement between the theory and experiment is less good. The reason for that is probably the hard-core approximation used in our calculations. In the ferromagnetic (spin-saturated) region one expects gapped magnon excitations in both spatially isotropic two-dimensional case and quasi-one-dimensional one.[1] This is why, the worse agreement between our theoretical approximate calculations and the experimentally observed data seems also natural. In Fig. 6 we present results for the best fit for the spatially anisotropic in-plane spin-spin interaction (quasi-one-dimensional case) and spatially isotropic in-plane case calculations for the sound attenuation as a function of the external magnetic field at $T = 500$ mK (which is higher than $T_N(B)$ for $B \gtrsim B_S$), compared with the experimental data. The agreement between the theoretical calculations and the experimental data seems satisfactory.

In conclusion, we performed low-temperature magnetoacoustic investigations of the quasi-two-dimensional spin system Cs_2CuCl_4. Our experimental data agree reasonably well with the results of a theoretical treatment of the sound attenuation and the change in the sound velocity, based on the spin-lattice interactions for low-dimensional spin-1/2 models. Our results can be considered as an additional support of the conclusions of those investigations for Cs_2CuCl_4,[8] which interpret the features in magnetic characteristics at $T > T_N$ and $B < B_s$ as the manifestation of the spin-liquid state. The obtained experimental data do not exclude the possibility of other interpretations. For example, the magnetic field effects can be taken into account within nonlinear spin-wave theory. Promising effort on this way have been done in Ref. 10 and announced in Ref. 21, but a self-consistent theoretical study, as far as we know, is still not completed.

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*Present address: London Centre for Nanotechnology, University College London, 17-19 Gordon Street, London WC1H 0AH, United Kingdom.

[1] See, e.g., A. A. Zvyagin, *Finite Size Effects in Correlated Electron Models: Exact Results* (Imperial College, London, 2005), and references therein.

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