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Magnetoacoustics of the Low-Dimensional Quantum Antiferromagnet Cs_2CuCl_4 with Spin Frustration

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Abstract We report on results of sound-velocity and sound-attenuation measurements in the triangular-lattice spin-$1/2$ antiferromagnet Cs_2CuCl_4 ($T_N = 0.6$ K), in external magnetic fields up to 14 T, applied along the b axis, and at temperatures down to 300 mK. The results are analyzed with a quasi-two-dimensional hard-core boson theory based on exchange-striction coupling. There is a good qualitative agreement between theoretical and experimental results.

Keywords Frustrated AFM · Hard-core bosons · Spin liquid · Magnetoacoustics

After the work of Anderson [1] spin-liquid quantum state has attracted a lot of interest. It is expected to exist especially in quasi-low-dimensional magnets, where

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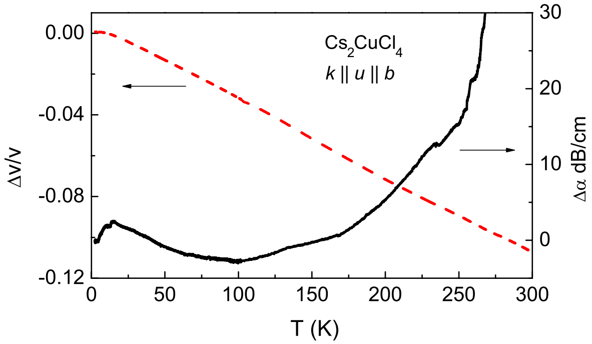


Fig. 1 (Color online) Temperature dependence of the relative change of the sound velocity (*left axis*) and sound attenuation (*right axis*) for the acoustic c_{22} mode ($k \parallel u \parallel b$) in zero applied magnetic field measured at 157 MHz

quantum and classical fluctuations, enhanced due to peculiarities in the densities of states, destroy magnetic ordering at any nonzero temperatures.

Cs_2CuCl_4 is considered as a candidate for spin-liquid behavior in the triangular $S = 1/2$ antiferromagnetic lattice. This new quantum phase is defined at zero temperature, but its effect can also be seen at higher temperatures [2]. For Cs_2CuCl_4 neutron scattering experiments [3, 4] indicated the existence of this phase. According to the neutron scattering, specific heat [5, 6], susceptibility, and magnetization [6] measurements, the long-range incommensurate antiferromagnetic order appears below $T_N = 0.6$ K. For the field applied along different crystallographic axes a number of magnetic phases have been observed [6]. Above $B_s(T = 0) \sim 8.9$ T, applied along b axis the saturated ferromagnetic phase appears. In the transition range $T_N < T \lesssim 2.6$ K the compound shows spin-liquid hallmarks [3]. Above 2.6 K, the magnetization obeys the Curie-Weiss law [6].

Cs_2CuCl_4 has an orthorhombic $Pnma$ crystal structure. The in-plane bc interactions $J = 4.34$ K along b and $J' = 0.34J$ along the zig-zag 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 [7].

Here, we present results on ultrasound investigations of Cs_2CuCl_4 carried out at temperatures down to 300 mK and in magnetic fields up to 14 T. We measured the relative change of the sound velocity and sound attenuation of the longitudinal acoustic c_{22}-mode ($k \parallel u \parallel [010]$, where k is the wave vector and u is the sound polarization) on a $3.6 \times 1.9 \times 2.8$ mm^3 sample of Cs_2CuCl_4, using a phase-sensitive detection technique based on a standard pulse-echo method with a set-up similar to the one described in [8]. In this way, the longitudinal acoustic wave modulates the largest exchange interaction J (exchange-striction coupling). The magnetic field was applied along the [010] direction, i.e., parallel to the b axis and to the sound-propagation direction. The absolute value of the sound velocity at room temperature has been determined as $v_l = (2620 \pm 20)$ m/s. The velocity increases by about 10% in the range from 300 K down to 300 mK (see Fig. 1).

At zero applied magnetic field the sound velocity grows linearly with decreasing temperature and does not reveal any anomalies down to low temperatures (see Fig. 2). In an applied field of about 6 T the temperature dependence of the sound velocity changes its character. Above 6 T the acoustic c_{22} mode reveals a softening below about 5 K and develops a fine structure around the Néel temperature of 0.62 K. This

Fig. 2 (Color online) Temperature dependence of the relative change of the velocity and attenuation of the acoustic c_{22}-mode in different magnetic fields probed with a longitudinal ultrasound signal at 42 MHz. The experimental geometry is $B \parallel k \parallel u \parallel b$

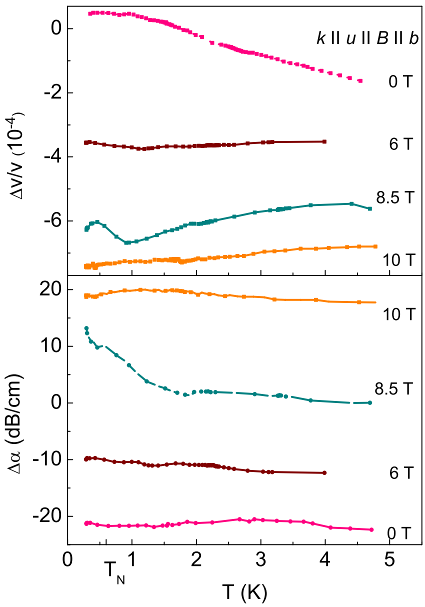
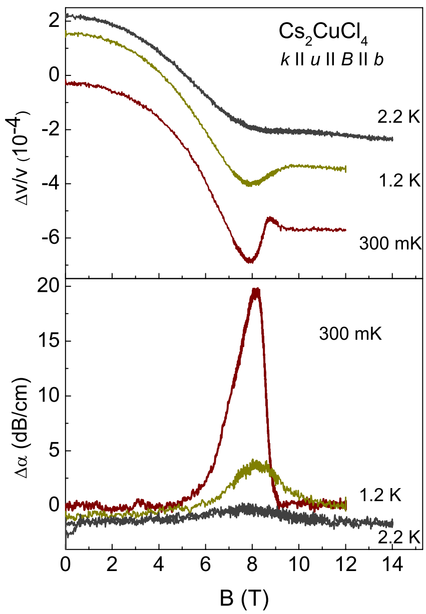


Fig. 3 (Color online) Field dependence of the relative change of the velocity and attenuation of the acoustic c_{22}-mode measured with an ultrasound signal at 42 MHz at different temperatures. The experimental geometry is $B \parallel k \parallel u \parallel b$



fine structure disappears in data taken at $B > 9$ T. For the sound velocity a small increase towards the lowest temperatures appears between about 6 and 8.5 T.

In Fig. 3, we present the field dependences of the acoustic properties measured at 42 MHz for different temperatures. The acoustic c_{22} mode shows a softening with increasing field and the sound velocity becomes constant in the fully polarized phase above 9 T. Below 1.5 K, the sound velocity has a dip anomaly around 8 T. For the data taken below 900 mK this minimum is followed by a step-like anomaly and a small maximum below 9 T. Thus, we observe a nonmonotonic behavior of sound velocity in the vicinity of the B_s [5, 6]. The sound attenuation exhibits a strong peak at about 8 T for temperatures below 1.2 K.

In magnetic materials the dominant contribution to the spin-lattice interactions mostly arises from the exchange-striction coupling. In our calculations we assumed that in Cs_2CuCl_4 the spatial dependence of the magnetic anisotropy is weaker than the spatial dependence of the exchange integrals. In this case, one can expect that only longitudinal sound waves interact with the spin subsystem. According to [9, 10], the relative renormalization of the longitudinal sound velocity can be written as

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

where

$$\begin{aligned} A_1 &= 2|G_0^z(\mathbf{k})|^2 \langle S_0^z \rangle^2 \chi_0^z + T \sum_{\mathbf{q}} \sum_{\alpha=x,y,z} |G_{\mathbf{q}}^\alpha(\mathbf{k})|^2 (\chi_{\mathbf{q}}^\alpha)^2, \\ A_2 &= H_0^z(\mathbf{k}) \langle S_0^z \rangle^2 + \frac{T}{2} \sum_{\mathbf{q}} \sum_{\alpha=x,y,z} H_{\mathbf{q}}^\alpha(\mathbf{k}) \chi_{\mathbf{q}}^\alpha. \end{aligned} \tag{2}$$

Here, N is the number of spins in the system, $\mathbf{q}$ is a 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 non-uniform magnetic susceptibilities, and the subscript 0 corresponds to $q = 0$. For spin systems with antiferromagnetic interactions, like Cs_2CuCl_4, the main contribution to the summation over $\mathbf{q}$ in (2) comes from terms with $q^{x,y,z} = \pi$. These magnetic characteristics were calculated in the framework of the hard-core boson theory for low-energy spin excitations for the spin subsystem of Cs_2CuCl_4. The renormalization is proportional to the spin-phonon coupling constants $G_{\mathbf{q}}^\alpha$ and $H_{\mathbf{q}}^\alpha$, which we used as fitting parameters [11]. The attenuation coefficient for Cs_2CuCl_4 reads as

$$\begin{aligned} \Delta\alpha (\equiv \Delta\alpha_k) = \frac{1}{Nv} \Bigg[& 2|G_0^z(\mathbf{k})|^2 \langle S_0^z \rangle^2 \chi_0^z \frac{\gamma_0^z}{(\gamma_0^z)^2 + \omega_{\mathbf{k}}^2} \\ & + T \sum_{\mathbf{q}} \sum_{\alpha=x,y,z} |G_{\mathbf{q}}^\alpha(\mathbf{k})|^2 (\chi_{\mathbf{q}}^\alpha)^2 \frac{2\gamma_{\mathbf{q}}^\alpha}{(2\gamma_{\mathbf{q}}^\alpha)^2 + \omega_{\mathbf{k}}^2} \Bigg], \end{aligned} \tag{3}$$

where $\gamma_{\mathbf{q}}^\alpha$ are the relaxation rates, which can be approximated by $\gamma_{\mathbf{q}}^\alpha = B/T\chi_{\mathbf{q}}^\alpha$, where B is a material-dependent constant (see [9]). In our calculations, we used the approximation, in which the relaxation rates do not depend on the direction and on the wave vector.

Our simplified theory reproduces the main features of the experimentally observed behavior: the pronounced minimum in the field dependences of the sound velocity and the peak in the attenuation at about B_s, the almost field-independent behavior of the acoustic characteristics at $B \gtrsim B_s$. The details will be published elsewhere [11].

In summary, our results demonstrate the important role which the magnetic excitations play in the vicinity of $T_N(B)$ and support the conclusions of previous investigations in Cs_2CuCl_4 [3, 4], which interpret the features in the behavior of the magnetic characteristics above the Néel temperature and for magnetic fields smaller than about B_s as the manifestation of the spin-liquid state.

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