Sound-wave anomalies in $SrCu_2(BO_3)_2$

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The temperature and high magnetic field dependence of the longitudinal sound-wave mode c_{11} in the two-dimensional dimer system $SrCu_2(BO_3)_2$ is presented. $c_{11}(T)$ shows anomalies due to strong interdimer spin-strain coupling. We can quantitatively interpret the temperature dependence of $c_{11}(T)$ together with the magnetic susceptibility $\chi_m(T)$ with a molecular field approximation of coupled dimer triplets. The sound velocity up to 50 T shows very sharp softening between the magnetization plateaus at low temperature. We argue that these pronounced effects arise from a resonant interaction between the phonons and the magnetic excitations which show softening between the plateaus.

The physics of low-dimensional spin systems is full of surprises. In zero magnetic field, one finds gapped ground states for antiferromagnetic Heisenberg chains with integer spins,[1] or for dimerized spin 1/2 systems (e.g., spin-Peierls systems[2]), or for spin 1/2 two-leg ladders,[3] to name just a few examples. Also, in the presence of a magnetic field instead of a smooth magnetization curve, one has found for some classes of materials[4] magnetization plateaus for rational fractions of the saturation magnetization. Examples given are the quasi-one-dimensional Ni compounds,[5] NH_4CuCl_3,[6] and the two-dimensional $SrCu_2(BO_3)_2$.[7]

This latter compound has a two-dimensional structure which consists of alternately stacked $CuBO_3$ and Sr layers. Within the $CuBO_3$ plane, the Cu^{2+}-Cu^{2+} dimers are orthogonally connected, giving rise to an exact orthogonal dimer ground state.[8] This system is topologically equivalent to the Shastry-Sutherland model.[9] In the inset of Fig. 1, two orthogonal dimers with the important exchange interactions J,J' are shown. Numerical evaluation of the corresponding Hamiltonian gives $J=100$ K, $J'=68$ K.[8,10,11] These authors also noted that the value $J'/J=0.68$ is very close to the value of a quantum phase transition (≈ 0.7).

In this paper we report the following features for this compound: (i) The longitudinal elastic constant c_{11} exhibits a strong temperature dependence which allows us to study the strain dependence and the elastic coupling between the Cu dimers. An important result of this analysis leads to an estimate for a possible structural phase transition. (ii) The sound velocity in high magnetic fields up to 50 T exhibits characteristic features related to the magnetization plateaus, very sharp minima before successive plateaus. A resonant interaction between the sound wave and the gap modes which soften between the plateaus might be responsible for these anomalies. This evidence is direct proof for the softening of the magnetic excitation between the plateaus. A theoretical treatment for these effects, especially a calculation of the magnetic excitation, is lacking so far.

In Figs. 1 and 2 we show the magnetic susceptibility and the elastic constant c_{11} as a function of temperature. Both thermodynamic quantities exhibit features typical for a low-dimensional spin gap system.

The magnetic susceptibility χ_m, previously published,[7,8] has a maximum at $T_{max}=18$ K and a strong decrease for lower temperatures indicative for a spin gap system. At high temperatures, a Curie-Weiss behavior with $\Theta=(J+4J')/4=93$ K is observed. For the following, it is interesting to note that a RPA-molecular field expression[12]

$$\chi_m=\chi_0/(1-j\chi_0) \tag{1}$$

gives a rather good fit for $T<300$ K. χ_0 is the magnetic susceptibility of an isolated dimer, a singlet-triplet system: $\chi_0=2e^{-\Delta/kT}/kTZ$ with Z the partition function. The fit gives a singlet-triplet gap for thermodynamic quantities $\Delta=29$ K rather close to the accepted value of 30 K,[8] and $j=-273$ K. The interdimer coupling is $j=4J'$ as expected.[8] Instead of starting with the Hamiltonian with the parameters J,J' (Ref. 8) we used directly the dimer susceptibility χ_0 with the renormalized value for $\Delta=29$ K$<J=100$ K.

In Fig. 2 we show the temperature dependence of the longitudinal constant c_{11}. For this mode the polarization and the propagation is along [100]. With the sound velocity v and the mass density $\rho=4.1$ g/cm^3 one gets $c_{11}=\rho v^2$. In Fig. 2(a) we plot $c_{11}(T)$ together with the temperature dependent background $c_{11}^0(T)$ taken from an empirical evaluation.[13] Similar to the magnetic susceptibility χ_m, we

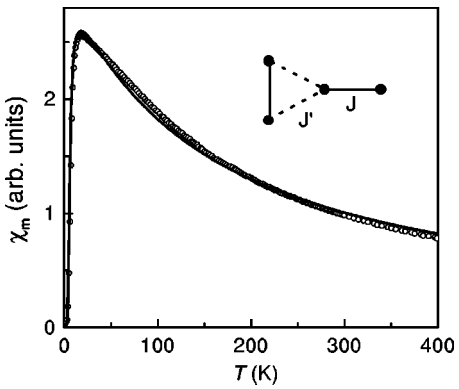

FIG. 1. Temperature dependence of magnetic susceptibility. Open circles: experiment from Ref. 7. Full line: fit with expression Eq. (1). Inset: two neighboring dimers with exchange interactions J,J'.

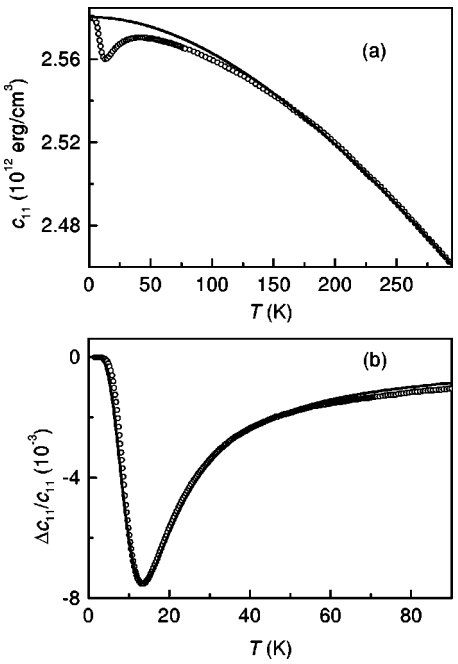

FIG. 2. (a) Temperature dependence of the c_{11} mode. Full line is background $c_{11}^0(T)$ as described in the text. The ultrasonic frequency was 98 MHz. (b) $(c_{11}(T)-c_{11}^0(T))/c_{11}^0(T=0)$ as a function of temperature. Open circles: experiment. Full line: fit from Eq. (2).

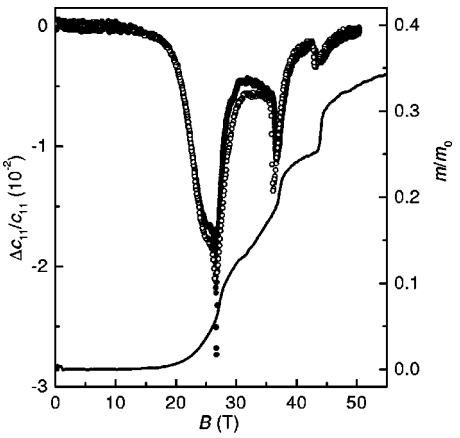

FIG. 3. Relative change of the elastic constant c_{11} for the magnetic field sweep up (solid circles) and sweep down (open circles) measured at $T=1.5$ K. Field dependence of relative magnetization m/m_0 at the same temperature is also shown (solid line). Here m_0 is a saturation value of the magnetization. The magnetic field is applied along the a axis. The elastic constant minima occur between the magnetization plateaus.

can interpret the temperature dependence of the elastic constant c_{11} with an analogous RPA expression.[14] In analogy to the magnetic susceptibility which is the response function of the magnetization to an applied field, we can define the strain susceptibility which is the response function to an applied strain.The elastic constant reads:

$$c_{11}(T)=c_{11}^0(T)-G^2\chi_{str} \quad \text{with} \tag{2}$$

$$\chi_{str}=\chi_s/(1-K\chi_s).$$

χ_s is the strain susceptibility of a single dimer,

$$\chi_s=G^{-2}\frac{\partial^2 F}{\partial\epsilon^2}=\frac{3e^{-\Delta/kT}}{kTZ^2}, \tag{3}$$

and K is the strength of the $(k=0)$ dimer-dimer interaction. The effective interaction K can be mediated, e.g., by phonons.

In Fig. 2(b) we compare $\Delta c_{11}/c_{11}=(c_{11}(T)-c_{11}^0(T))/c_{11}^0(T=0)$ with a fit to χ_{str} using the same $\Delta=29$ K. Here the minimum of the elastic constant is at $T_{min}=13$ K and the coupling constant $K=23.8$ K. The fit is also very good. This analysis shows that with a three times larger coupling constant K, a structural transition would take place. The single dimer coupling constant $G=\partial\Delta/\partial\epsilon$ which measures the strain dependence of the singlet-triplet gap amounts to $G\approx 900$ K [with $c^0(T=0)=25.8\times10^{11}$ erg/cm^3 at low temperature, and the number of dimers $n=0.74\times10^{22}$ cm^{-3}). So this simple RPA expressions account correctly for the T_{max} and T_{min} in χ_m and χ_{str}, respectively. It also gives a good overall fit for χ_m and c_{11} using the same singlet-triplet gap. It further shows that the strain interactions are very important for this dimer compound.

Now we proceed to the magnetic field dependence. In Fig. 3 we show the relative change of the same elastic constant c_{11} as a function of magnetic field up to 50 T at 1.5 K. The ultrasonic technique recently developed for pulsed fields is described elsewhere.[15] In the present case the rise time of the pulse is 8 ms and the total pulse duration is 24 ms. In Fig. 3 both curves for field up and down are shown. No substantial heating effects are discernible, we estimate it to $\Delta T<0.5$ K. For 1.5 K we observe a broad minimum at 25 T followed by very sharp minima at 27 T, 36 T, and 42 T. In this figure the magnetization for the same temperature is also included. It clearly exhibits magnetization plateaus for $m/m_0=1/8$, 1/4, and 1/3. It is seen that the sound velocity minima are in the region where the magnetization changes from one plateau to the next one.

The broad minimum at 25 T arises possibly from localized triplet excitations. Existence of a strong spin-phonon coupling was pointed out recently by Ueda and Miyahara.[16] If the spin-phonon coupling is strong enough there is the possibility of the formation of a self-trapped triplet, a bound complex of triplet excitation, and local distortion (triplet-phonon-bound state). The energy of the self-trapped triplet excitations is lower than the usual triplet branch, which may explain this broad minimum.

The first strong softening at 27 T is due to the interaction between the sound wave and the soft triplet branch. Inelastic neutron scattering determined $\Delta=35$ K.[17] This leads to a softening at 27 T with $g=2$, in agreement with our observation. We observed the same effect recently in $(VO)_2P_2O_7$.[18] The evolution of this sound velocity minimum with temperature is similar in both substances: Comparing $\Delta c_{11}/c_{11}$ for 1.7 K and 7.4 K, we observe a small shift from 26.7 T to 27.4 T and a broadening with increasing temperature. This is seen in Fig. 4 where the field dependence of the relative elastic constant is plotted for different temperatures. The sharp minima between the plateaus disappear rapidly with increasing temperature, and at 7.4 K only the first minimum survives.

We can estimate again the coupling constant $\partial\Delta/\partial\epsilon$ from

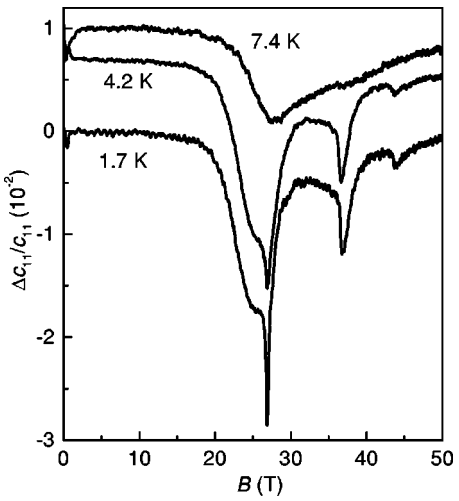

FIG. 4. Relative change of the elastic constant c_{11} versus magnetic field at three different temperatures. Only field sweeps up are shown. The curves were shifted arbitrarily for the sake of clarity.

the softening of the first triplet at 27 T where $\Delta c_{11}/c_{11} \approx 2.8\%$. Since the first magnetic excitation shows no dispersion[17] we can again use the free energy of weakly coupled dimers. We get $\partial\Delta/\partial\epsilon \approx 700$ K in rough agreement with the estimate from the c_{11} temperature dependence given above. This indicates that both effects are due to an exchange striction coupling.

Now we discuss the additional steep minima effects at 36 T and 42 T, the steep minima before each plateau ($m/m_0 = 1/4, 1/3$). We argue that these are likewise resonant effects between the sound wave and the corresponding soft mode before the plateau is reached. Within the plateau the excitation modes have a gap, thus keeping the magnetization constant. While we are not aware of any theory describing these excitation modes and their interaction with the elastic modes, we can give the field dependence of our sound velocities between the plateaus: $\Delta c_{11}/c_{11} \propto (B - B_c)^{\gamma}$ with $\gamma \approx 0.5$ for the resonances before the plateaus for $B > B_c$ at $m/m_0 = 1/4, 1/3$.

In summary, we have given temperature and field dependencies of a longitudinal elastic constant in the two-dimensional compound $SrCu_2(BO_3)_2$. We find considerable phonon induced dimer-dimer coupling and a strain dependence of the singlet-triplet gap of 700–900 K in rough agreement with exchange striction coupling constants ($\partial J/\partial\epsilon \approx -9J$).[14] The sharp softening of c_{11} between the plateaus indicates a resonant interaction with the magnetic excitation spectrum which softens between the plateaus. Further investigations of other elastic modes can help in elucidating the strain-magnetic excitation interaction in this material. It is clear that phonon effects are very important in this compound. A more detailed theoretical treatment of this interaction and of the magnetic excitations is needed.

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[1] F. D. M. Haldane, Phys. Lett. **93A**, 464 (1983); Phys. Rev. Lett. **50**, 1153 (1983).
[2] For a review, see J. P. Boucher and L. P. Regnault, J. Phys. I **6**, 1939 (1996).
[3] For a review of ladder systems, see E. Dagotto and T. M. Rice, Science **271**, 618 (1996).
[4] M. Oshikawa, M. Yamanaka, and I. Affleck, Phys. Rev. Lett. **78**, 1984 (1997); K. Totsuka, Eur. Phys. J. B **5**, 705 (1998).
[5] R. Sato *et al.*, J. Magn. Magn. Mater. **177-181**, 645 (1998).
[6] W. Shiramura *et al.*, J. Phys. Soc. Jpn. **67**, 1548 (1998).
[7] H. Kageyama *et al.*, Phys. Rev. Lett. **82**, 3168 (1999).
[8] S. Miyahara and K. Ueda, Phys. Rev. Lett. **82**, 3701 (1999).
[9] B. S. Shastry and B. Sutherland, Physica B & C **108B**, 1069 (1981).
[10] S. Miyahara and K. Ueda, Phys. Rev. B **61**, 3417 (2000).
[11] Z. Weihong *et al.*, Phys. Rev. B **60**, 6608 (1999).
[12] R. M. White, *Quantum Theory of Magnetism* (Springer, Berlin, 1983).
[13] Y. P. Varshni, Phys. Rev. B **2**, 3952 (1970).
[14] B. Lüthi, in *Dynamical Properties of Solids*, edited by G. K. Horton and A. A. Maradudin (North-Holland, Amsterdam, 1980).
[15] B. Wolf *et al.*, Europhys. Lett. **48**, 182 (1999).
[16] K. Ueda and S. Miyahara (unpublished).
[17] H. Kageyama *et al.* (unpublished).
[18] B. Wolf *et al.*, J. Appl. Phys. **87**, 7055 (2000).