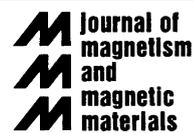




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# Ultrasonic experiments in $\text{SrCu}_2(\text{BO}_3)_2$ and $\text{NH}_4\text{CuCl}_3$ in magnetic fields up to 50 T

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## Abstract

We present sound velocity data in high fields for the two plateau substances  $\text{SrCu}_2(\text{BO}_3)_2$  and  $\text{NH}_4\text{CuCl}_3$ . Pronounced sound velocity and attenuation anomalies are observed in the region between the magnetization plateaus. We interpret these as a resonant interaction between the sound wave mode and the soft magnetic excitations. © 2001 Elsevier Science B.V. All rights reserved.

**Keywords:** Low dimensional spin system; Magnetization plateau

Low dimensional spin systems exhibit many surprising properties. For example, dimerised spin chains or two dimensional spin dimers exhibit distinct plateaus in magnetization as a function of magnetic field. In  $\text{SrCu}_2(\text{BO}_3)_2$  [1] and in  $\text{NH}_4\text{CuCl}_3$  [2] these have been measured for various temperatures.

With a quadrature method [3] we are able to measure ultrasonic velocity and attenuation in pulsed fields up to 50 T. We applied this technique to various substances with interesting magnetic properties:  $\text{CsCuCl}_3$  [3],  $\text{YbInCu}_4$  [4],  $(\text{VO})_2\text{P}_2\text{O}_7$  [5],  $\text{SrCu}_2(\text{BO}_3)_2$  [6] and  $\text{NH}_4\text{CuCl}_3$  [7]. Here we briefly discuss the latter two substances which exhibit plateaus in the magnetization.

$\text{SrCu}_2(\text{BO}_3)_2$  has a two dimensional structure which consists of alternately stacked  $\text{CuBO}_3$ - and Sr-layers. Within the  $\text{CuBO}_3$  plane the  $\text{Cu}^{2+}$  form dimers, which are orthogonally connected giving rise to an exact orthogonal dimer ground state [8]. In Fig. 1 we show a longitudinal sound velocity ( $c_{11}$  mode) versus magnetic field

for  $T = 1.5$  K. The magnetization for the same temperature is also given in the same figure. We observe for the relative elastic constant a broad minimum at 25 T followed by very sharp minima at 27, 36, and 42 T. The corresponding magnetization exhibits plateaus for  $m/m_0 = \frac{1}{8}$ ,  $\frac{1}{4}$  and  $\frac{1}{3}$ . 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 can arise from localized triplet excitations (triplet-phonon-bound state). The sharp minima for higher fields arise due to a resonant interaction between the magnetic excitations and the soundwave mode.

The interaction between soundwaves and a triplet magnetic state was investigated experimentally in the chain substance  $(\text{VO})_2\text{P}_2\text{O}_7$  [5]. The coupling between a longitudinal sound mode and the lowest branch of the triplet in a magnetic field is due to the exchange striction mechanism. This leads to a strain dependence of the singlet-triplet splitting. A mean field type description gives the salient features of the field and temperature dependence of the sound velocity for this substance [5].

From the temperature dependence of the  $c_{11}$  elastic mode and from the resonant soundwave-spin triplet excitation we get a magnetoelastic coupling constant of ca

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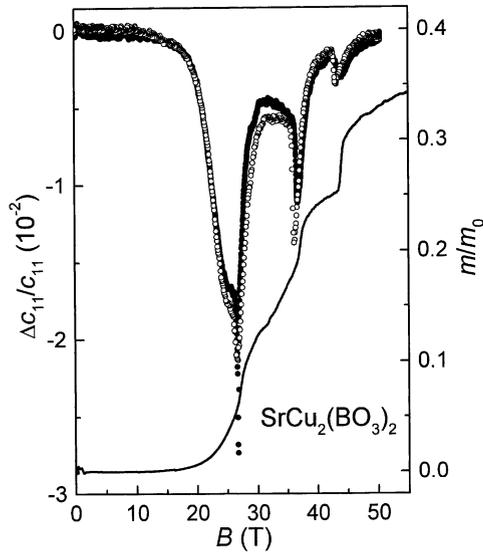


Fig. 1. Relative change of the elastic constant  $c_{11}$  in  $\text{SrCu}_2(\text{BO}_3)_2$  for the magnetic field sweep up (solid circles) and sweep down (open circle) measured at  $T = 1.5$  K. Field dependence of relative magnetization  $m/m_0$  at the same temperature is also shown (solid line). Here  $m_0$  is a saturation value of the magnetization. The magnetic field applied along the  $a$  - axis. The elastic constant minima occur between the magnetization plateaus.

800 K for  $\text{SrCu}_2(\text{BO}_3)_2$  [6] This large coupling constant means that the Cu-dimers are coupled very strongly to the lattice. But it is not large enough to induce a structural transition. Note that the exchange constants are close to a critical value to induce an antiferromagnetic phase transition [8]. Further studies with other elastic modes are necessary to gain a complete picture of the strain-dimer coupling.

$\text{NH}_4\text{CuCl}_3$  belongs to the  $\text{ABX}_3$  family of compounds which plays a prominent role in low dimensional magnetism.  $\text{KCuCl}_3$ ,  $\text{TlCuCl}_3$  and  $\text{NH}_4\text{CuCl}_3$  have a monoclinic structure at room temperature [9].  $\text{NH}_4\text{CuCl}_3$  has a structural order-disorder type transition at 70 K. Several elastic modes exhibit pronounced anomalies in the vicinity of this transition [7].

In Fig. 2 we show elastic constant results for  $\text{NH}_4\text{CuCl}_3$ . The shear mode  $c_{66}$  is plotted as a function of magnetic field at 1.5 K. Pronounced anomalies are observed at the onset and the end of the plateau states i.e., for 4.5, 13, 17, 24 and 28 T. One can compare the results in Fig. 2 with magnetization and differential susceptibility results for this substance [2]. The similarity of the relative soundvelocity results with the negative susceptibility is remarkable. In both cases the effect between the  $\frac{1}{4}$  and  $\frac{3}{4}$  plateau is the largest. Note that for  $\text{SrCu}_2(\text{BO}_3)_2$  such a similarity does not hold. For the field up and the field down curve we notice some hysteresis in the magni-

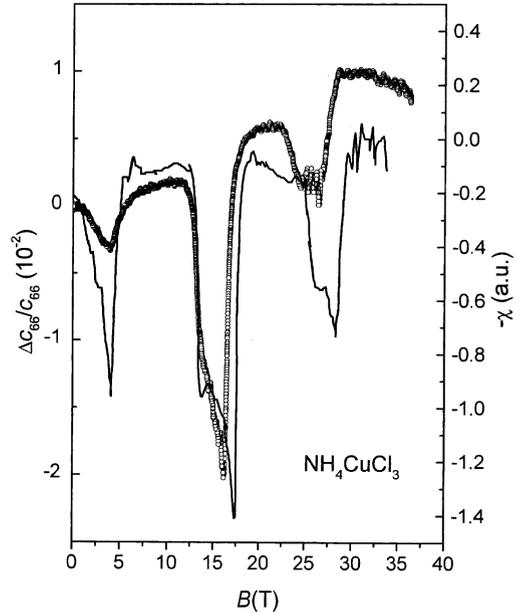


Fig. 2. Magnetic field dependence of the transversal mode  $c_{66}$  together with the magnetic susceptibility from Ref. [9]. (solid line) for  $\text{NH}_4\text{CuCl}_3$ . Both measured at  $T = 1.5$  K. For the elastic constant only the magnetic field sweep down (open circles) is shown. The external magnetic field is applied parallel to the  $a$  - axis.

tude (not shown). We do not know yet whether this is an intrinsic effect. As in the case of  $\text{SrCu}_2(\text{BO}_3)_2$  the sound wave anomalies arise due to a resonant interaction with the soft magnetic mode between the different plateaus. The size of the effect for the  $c_{66}$  mode is similar in size with the longitudinal mode for  $\text{SrCu}_2(\text{BO}_3)_2$  shown in Fig 1. The longitudinal mode  $c_{22}$  for  $\text{NH}_4\text{CuCl}_3$  exhibits almost an order of magnitude smaller effect in the field dependence [7].

Various magnetic properties for  $\text{NH}_4\text{CuCl}_3$  are not understood. Contrary to the related compounds  $\text{KCuCl}_3$  and  $\text{TlCuCl}_3$  one observes no singlet-triplet gap for  $B = 0$  [9]. In addition the absence of a plateau for  $m/m_0 = \frac{1}{2}$  is a mystery. This sound wave-magnetic excitation effect allows to describe the spin lattice coupling in this substances more completely.

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## References

- [1] H. Kageyama, K. Yoshimura, R. Stern, N.V. Mushnikov, K. Onizuka, M. Kato, K. Kosuge, C.P. Slichter, T. Goto, Y. Ueda, Phys. Rev. Lett. 82 (1999) 3168.

- [2] W. Shiramura, K. Takatsu, B. Kurniawan, H. Tanaka, H. Uekusa, Y. Ohashi, K. Takizawa, H. Mitamura, T. Goto, *J. Phys. Soc. Japan* 67 (1998) 1548.
- [3] B. Wolf, S. Zherlitsyn, S. Schmidt, B. Lüthi, *Europhys. Lett.* 48 (1999) 182.
- [4] S. Zherlitsyn, B. Lüthi, B. Wolf, J.L. Sarrao, Z. Fisk, V. Zlatic, *Phys. Rev. B* 60 (1999) 3148.
- [5] B. Wolf, S. Schmidt, H. Schwenk, S. Zherlitsyn, B. Lüthi, *J. Appl. Phys.* 87 (2000) 7055.
- [6] S. Zherlitsyn, et al., to be published.
- [7] S. Zherlitsyn et al., to be published.
- [8] S. Miyahara, K. Ueda, *Phys. Rev. Lett.* 82 (1999) 3701.
- [9] H. Tanaka, W. Shiramura, T. Takatsu, B. Kurniawan, M. Takahashi, K. Kamishima, K. Takizawa, H. Mitamura, T. Goto, *Physica B* 246–247 (1998) 230.