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Lattice Instabilities in the Frustrated Magnet $CdCr_2O_4$: An Ultrasonic Study

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Abstract We report results on ultrasound studies of the frustrated magnet $CdCr_2O_4$. This compound demonstrates an antiferromagnetic ordering at $T_N = 7.8$ K and a metamagnetic phase transition at 28 T followed by a wide magnetization plateau with one half of the full moment of $S = 3/2$ of Cr^{3+} ions. A longitudinal acoustic mode in the [111] crystallographic direction exhibits pronounced effects both in the temperature and magnetic-field dependencies. Pulsed-field measurements show a drastic change in the sound velocity just below and above the 1/2 magnetization plateau. Our results suggest a large spin-strain coupling and give evidence for a pronounced interplay between spin and lattice degrees of freedom in $CdCr_2O_4$.

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Frustrated spin systems with nearest-neighbor interaction attract much attention because of the high degeneracy of the ground state and the tendency to form exotic phases. A realization of such systems in three dimensions (3D) is the pyrochlore lattice, consisting of a network of corner-sharing tetrahedra with spins sitting at the vertices of the corner. A typical example is the spinel compound $CdCr_2O_4$ where Cr^{3+} ions with spin $S = 3/2$ build the 3D pyrochlore lattice. In this compound, the Curie-Weiss temperature $\theta = -70$ K is an order of magnitude larger than the Neel temperature indicating a high level of magnetic frustration [1]. The antiferromagnetic (AF) ordering in $CdCr_2O_4$ at $T_N = 7.8$ K is accompanied by a cubic ($Fd\overline{3}m$) to tetragonal ($P4_332$) structural change, which can be considered as a spin Jahn-Teller phase transition, driven by spin frustrations. The spin-orbit coupling is negligible in this compound. Neutron-diffraction studies [2, 3] revealed the presence of a spiral spin structure below T_N, which transforms to a four-sublattice canted structure at a magnetic field of 5.7 T as indicated by ESR measurements [4]. Magnetization data exhibit a broad plateau between 28 and about 60 T [1, 5], which corresponds to $1/2$ of the saturation magnetization. The fully polarized state is achieved above 90 T in this system [5]. Only a small dependence on different field orientations has been observed [1]. $CdCr_2O_4$ can be described using the nearest-neighbor Heisenberg model with AF exchange interactions. Theoretical studies of the spin and crystallographic structures at high magnetic field have been reported in Refs. [6–8]. The important role of the spin-lattice interactions in $CdCr_2O_4$ has been underlined in a number of works [1–3, 6–8].

A powerful tool to probe spin-lattice interactions and lattice instabilities is given by ultrasound measurements [9]. This technique is available for non-destructive pulsed magnetic fields, extending the parameter space in modern material research. We have performed measurements of the relative change of the sound velocity and the sound attenuation for longitudinal acoustic c_L mode propagating along the [111] direction of $CdCr_2O_4$. This acoustic mode corresponds to $c_L = 1/3(c_{11} + 2c_{12} + 4c_{44})$ elastic constants for a cubic crystal. The magnetic field was applied along the [111] direction, i.e., parallel to the wave vector $\mathbf{k}$ and the polarization $\mathbf{u}$ of the ultrasound wave. The single crystal was grown using a flux method. The sample thickness along the [111] direction is 0.83 mm.

Figure 1 shows the temperature dependence of the sound velocity and the attenuation of the c_L mode measured below 240 K at zero magnetic field. The c_L mode exhibits a softening below 120 K followed by a minimum at approximately 13 K and a jump-like anomaly at T_N, which is accompanied by a small hysteresis (not shown). The gradual decrease of c_L occurs at temperatures corresponding to a cooperative paramagnetic state of the Heisenberg pyrochlore antiferromagnet. In this regime the change of the sound velocity is proportional to the nearest-neighbor spin correlations $\langle S_i S_j \rangle$ and can be successfully modeled by Monte Carlo simulations (to be published). Note, that the infrared reflectivity spectra exhibit a phonon softening in the same temperature range [10, 11]. The hysteresis in the sound velocity at T_N confirms the first-order type of the phase transition, previously suggested from magnetic-susceptibility measurements [1]. The sound attenuation increases when lowering the

Fig. 1 (Color online) Change of the sound velocity (*blue curve*) and the sound attenuation (*red curve*) versus temperature for the c_L mode in $CdCr_2O_4$ for an ultrasound frequency of 107 MHz

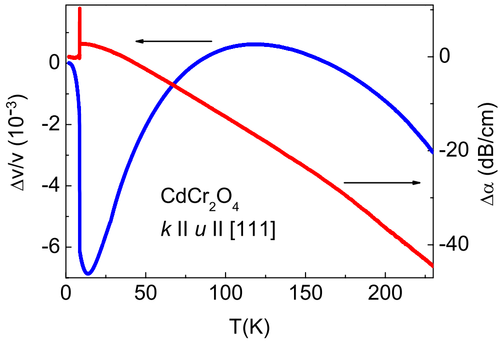
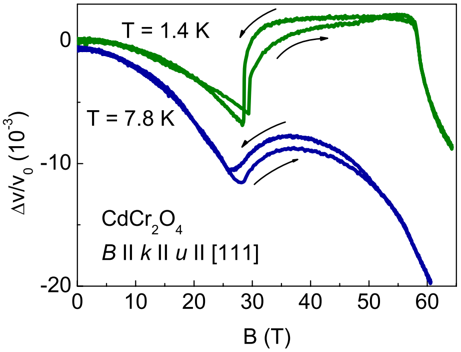


Fig. 2 (Color online) Change of the sound velocity as a function of magnetic field for the c_L mode in $CdCr_2O_4$ measured at 1.4 K (*green curve*) and 7.8 K (*blue curve*) for an ultrasound frequency of 81 MHz. The experimental geometry is $B \parallel k \parallel u \parallel [111]$



temperature and demonstrates a peak-like anomaly at T_N. The attenuation level in the ordered phase is lower than at temperatures slightly above the phase transition. The strong anomalies in the acoustic properties reflect the crucial role of the spin-strain coupling in this compound.

Applied magnetic fields affect the lattice degrees of freedom through these strong spin-strain interactions. We have performed pulsed magnetic-field experiments in $CdCr_2O_4$ in order to study the acoustic behavior. Results for the sound velocity in magnetic fields up to 63 T at temperatures of 1.4 and 7.8 K are shown in Fig. 2. At 1.4 K, the sound velocity decreases first, then demonstrates a jump at the magnetic field where the magnetization plateau appears. This anomaly corresponds to a first-order phase transition from the spiral AF structure to a collinear spin configuration with three spins up and one spin down at each Cr^{3+} tetrahedra. Previously, a large magnetostriction has been reported at this phase transition [1]. A cubic crystallographic structure has been suggested from high-field x-ray experiments at the plateau state [12]. It has been proposed [6] that the lattice distortion stabilizes the 3-up 1-down collinear spin configuration. Indeed, there is only a slight change of the sound velocity within the magnetization-plateau range. The plateau terminates at approximately 58 T, what is confirmed by a sharp anomaly, i.e., an abrupt decrease in the sound velocity. This anomaly corresponds to a phase transition to a non-collinear canted spin configuration [5, 6]. It is interesting to note that the hysteresis in the sound velocity takes place not only at the first-order phase transition around 28 T but it spreads along the whole plateau range up to 58 T, showing a complicated interplay between the spin and lattice degrees of freedom within the magnetization plateau. The sound velocity change, which takes place between 58 and 63 T, is even larger than the anomaly at 28 T. The highest applied magnetic field of 63 T is not sufficient to detect the complete sound velocity change. No hysteresis has been detected at 58 T, hinting at a second-order type of this phase transition. It is worth to note that the magnetization exhibits only a smooth, kink-like anomaly at this phase transition [5]. A transverse spin order which is equivalent to a Bose-Einstein condensation of magnons is predicted from quantum-fluctuation theory at magnetic field just above the plateau [8].

The first order phase transition at about 28 T could be resolved by magnetization measurements up to temperatures slightly above T_N [1]. Our pulsed-field ultrasound measurements performed at 7.8 K (Fig. 2) also clearly reveal an anomaly in the sound velocity at this field. The anomaly at 28 T is somewhat smoother than at lower temperatures, but still clearly evident. The hysteresis survives and the total change in the sound velocity is even larger at higher temperatures approaching 2% between zero and 60 T.

The important question, which is raised now, concerns the symmetry and strength of the magneto-elastic coupling in $CdCr_2O_4$. This question demands adequate models and a thorough analysis of the experimental results. We would like to mention that in the current investigation we deal with the acoustic c_L mode where various deformations are involved (see the c_L mode definition above). This fact complicates a symmetry analysis of the obtained data. In addition, below T_N, in the tetragonal phase, there are three types of domains corresponding to an elongation of the *c*-axis, and all of them contribute to the acoustic c_L mode.

In summary, we have presented a magneto-acoustic study of the frustrated spin system $CdCr_2O_4$. Strong sound velocity anomalies have been observed at the magnetic phase transitions in $CdCr_2O_4$. The spin-strain coupling is crucial and determines the underlying physics of this compound, reducing a geometric frustration effect and lifting the degeneracy in the system. The spin-strain coupling has an exchange-striction character in $CdCr_2O_4$; a detailed analysis will be published elsewhere.

Acknowledgements This research was partly supported by EuroMagNET II under EU contract 228043.

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