Spin-strain coupling in $NiCl_2$-$4SC(NH_2)_2$

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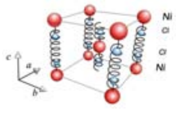
Motivation

- Direct probing of spin-phonon interaction
- Study the influence of quantum critical points on the phonon degrees of freedom
- Experimental test for the magnetic spin models of low-dimensional magnets

Structure and properties of **D**ichloro-*tetrakis*-**T**hiourea-**N**ickel (II), or **DTN**

- Body-centered tetragonal crystal structure
- Ni^{2+}: $S = 1$ spin-chains along the c axis
- Single-ion anisotropy D = 8.9 K; exchange parameters: intra-chain J_c = 2.2 K; inter-chain $J_{a,b}$ = 0.18 K [2,3]
- Gap in magnetic excitations dominated by D



Simplified structure of DTN: the S = 1 spins of the Ni ions form chains along the c direction.
From Ref. 4.

- There are two quantum critical points: $H_c \approx$ 2.1 T and $H_s \approx$ 12.6 T
- For T_N^{max} < 1.2 K and $H_c < H < H_s$: 3D long-range order (canted XY-AFM) [1,2]
- Magnetostriction along the spin-chains: $\Delta L/L$ < 10^{-4} [4]

Experimental results

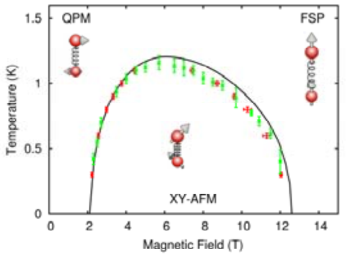


Fig. 1: Phase diagram of DTN for magnetic fields parallel to the c direction. The green and red dots are obtained from the ultrasonic measurements presented here, the solid line depicts results from Ref. [3]. The inserted drawings depict the model from Ref. [4].

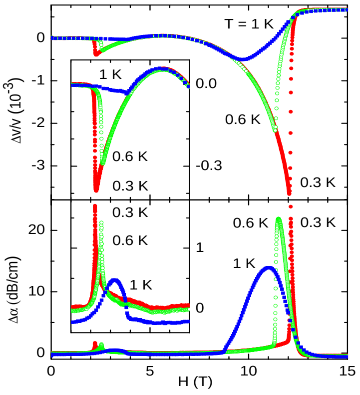


Fig. 2: Relative change of sound velocity and attenuation of the c_{33}-mode at 78 MHz, as a function of magnetic field along the c axis for $T < T_N^{max}$. The insets show the curves close to H_c with an enlarged scale.

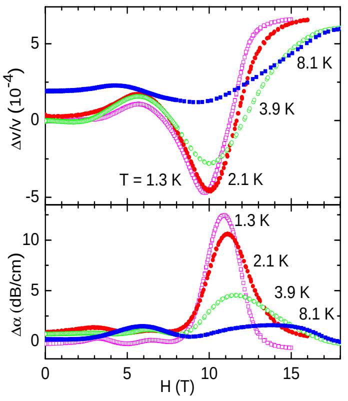


Fig.3: Relative change of sound velocity and attenuation of the c_{33}-mode at 78 MHz as a function of magnetic field parallel to the c axis for $T > T_N^{max}$.

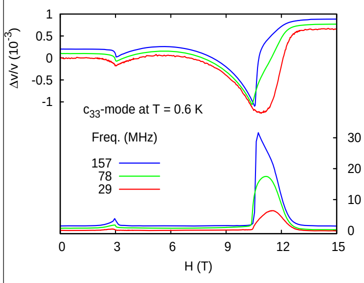


Fig. 4: Relative change of sound velocity and attenuation of the c_{33}-mode as a function of magnetic field, at T = 0.6 K and for three frequencies: 29, 78, and 157 MHz. The curves are vertically offset for clarity.

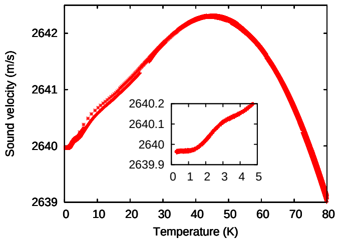
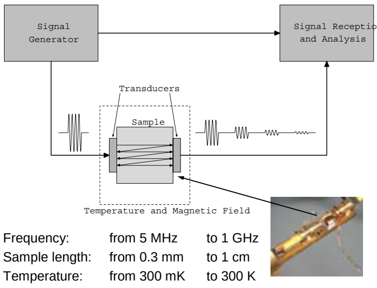


Fig. 5: Sound velocity of the c_{33}-mode at ~78 MHz in DTN as a function of temperature. Data were obtained in two different cryostats.

Experimental details

- We use a phase-sensitive detection technique based on a standard ultrasonic pulse-echo method.
- The relative changes of the velocity and attenuation of longitudinal sound wa-ves propagating along the c axis (c_{33}-mode) are measured as a function of temperature T and external magnetic field H (applied along the c axis).
- Absolute measurements of the sound velocity at room temperature and at low temperatures were performed for different frequencies.



Frequency:	from 5 MHz	to 1 GHz
Sample length:	from 0.3 mm	to 1 cm
Temperature:	from 300 mK	to 300 K
Magnetic field:	from 0 T	to 20 T
Resolution:	$\Delta v/v \sim 10^{-6}$	$\Delta\alpha \sim 10^{-3}$

- Strong spin-lattice interaction was observed both in the disordered and ordered phases (Fig. 2 and 3).
- Strong frequency dependence of anomalies in the critical region was observed (Fig. 4).
- The anomalies allow to map the phase diagram, which is in agreement with results from Ref. [2] and [3] (Fig. 1).
- The sound velocity shows a maximum at ~44 K (Fig. 5).

Discussion

- The spin-phonon interaction can be explained in terms of exchange striction.
- The effective free-fermion theory yields a good qualitative description for the behavior of the sound velocity and attenuation [5]. (Fig. 6)
- The frequency dependence in the critical regions indicates that relaxation processes are relevant.

Theory

- Effective free-fermion theory was applied to the 1D S = 1 spin chain in the gaped regime.
- E.g., the sound velocity change reads as

$$\frac{\Delta v_l}{v_l} = -\frac{A_1 + A_2}{(N\omega_k)^2},$$
$$A_1 = 2|g_0(k)|^2 \left\langle S_0^z \right\rangle^2 \chi_0^z + T\sum_q \sum_{\alpha=x,y,z} |g_q^\alpha(k)|^2 (\chi_q^\alpha)^2,$$
$$A_2 = |h_0(k)|^2 \left\langle S_0^z \right\rangle^2 + \frac{T}{2}\sum_q \sum_{\alpha=x,y,z} h_q^\alpha(k)(\chi_q^\alpha).$$

where g_q^α and h_q^α - spin-phonon coupling constants;
$\chi_q^\alpha = \chi_q^{\alpha(1)}/[1 - ZJ_{ab}\chi_q^{\alpha(1)}]$ – magnetic susceptibility with Z – the coordination number, $\chi_q^{\alpha(1)}$ – susceptibility of 1 chain

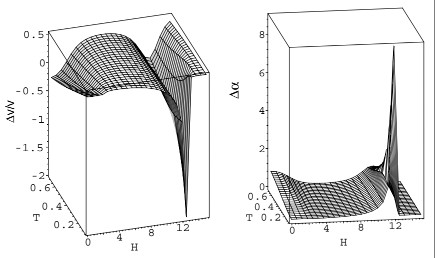


Fig. 6: Relative change of velocity (left) and attenuation (right) of longitudinal sound as a function of temperature and magnetic field, calculated within the framework of the effective free-fermion theory.

Conclusions

- There is a strong spin-phonon interaction in DTN.
- The sound velocity and the attenuation are renormalized in the vicinity of the quantum critical points.
- The (H,T) phase diagram was determined with high accuracy.

Outlook

- Clarify the origin of the sound-velocity maximum at ~44 K.
- Analysis of the relaxation processes in the critical region

References

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