The temperature dependence of phonon specific heat in Cu(en)Cl was studied up to 300 K in zero magnetic field. The analysis of the contribution of acoustic modes performed within Debye approximation yielded Debye temperature $\theta_D = 95.076^\circ$. The red line represents total phonon specific heat.

The energy of the IR modes in Cu(en)Cl was studied up to 300 K in zero magnetic field. The analysis of the contribution of acoustic modes performed within Debye approximation yielded Debye temperature $\theta_D = 95.076^\circ$. The red line represents total phonon specific heat.

Measurements of the temperature dependence of the heat capacity were performed with the Quantum Design Physical Property Measurement System (PPMS) at the Institute of Physics, Slovak Academy of Sciences, Košice, Slovakia. The measurements were carried out in the temperature range from 1.8 to 300 K in a zero magnetic field for single crystal samples.

The measurement of Raman spectra was performed using a Bruker vacuum FTIR spectrometer, model VERTEX 80 v with a RAM II module for measuring Raman spectra at the Institute of Experimental Physics of the Slovak Academy of Sciences in Košice, which has a working range from 50 to 3500 cm$^{-1}$. Experiments were performed at room temperature on powder samples placed in an aluminum capsule. The power of the laser with a wavelength $\lambda = 1064$ nm was 10 mW and reproducible vibrational spectra were obtained for the number of scans 2500.

**Experimental Results**

**Cu(en)Cl$_2$**

- Raman spectrum of a polycrystalline Cu(en)Cl$_2$ sample measured at room temperature. Arrows indicate frequencies of IR active modes (G. Paliani et al., Chem. Phys. 4, 440 (1974)).

- Raman spectrum of a polycrystalline Cu(en)Cl$_2$ sample measured at room temperature. Arrows indicate frequencies of IR active modes (J. Chem. Crystallogr. 21, 15 (1994)).

**Cu(tn)Cl$_2$**

- Raman spectrum of a polycrystalline Cu(tn)Cl$_2$ sample measured at room temperature. Arrows indicate frequencies of IR active modes (J. Chem. Crystallogr. 21, 15 (1994)).

- Raman spectrum of a polycrystalline Cu(tn)Cl$_2$ sample measured at room temperature. Arrows indicate frequencies of IR active modes (J. Chem. Crystallogr. 21, 15 (1994)).

**Cu(en)(H$_2$O)$_2$SO$_4$**

- Raman spectrum of a polycrystalline Cu(en)(H$_2$O)$_2$SO$_4$ sample measured at room temperature. Arrows indicate frequencies of IR active modes (J. Chem. Crystallogr. 21, 15 (1994)).

- Raman spectrum of a polycrystalline Cu(en)(H$_2$O)$_2$SO$_4$ sample measured at room temperature. Arrows indicate frequencies of IR active modes (J. Chem. Crystallogr. 21, 15 (1994)).

**Cu(en)$_2$SO$_4$**

- Raman spectrum of a polycrystalline Cu(en)$_2$SO$_4$ sample measured at room temperature. Arrows indicate frequencies of IR active modes (J. Chem. Crystallogr. 21, 15 (1994)).

- Raman spectrum of a polycrystalline Cu(en)$_2$SO$_4$ sample measured at room temperature. Arrows indicate frequencies of IR active modes (J. Chem. Crystallogr. 21, 15 (1994)).

Conclusions

- The temperature dependence of specific heat in Cu(en)Cl, Cu(tn)Cl, Cu(en)$_2$SO$_4$, and Cu(en)(H$_2$O)$_2$SO$_4$ was studied up to 300 K in zero magnetic field.

- The analysis of the contribution of acoustic modes performed within Debye approximation yielded Debye temperature $\theta_D = 147$ K, 109 K, and 93 K for Cu(en)$_2$SO$_4$, Cu(en)Cl$_2$, Cu(en)$_2$SO$_4$, and Cu(en)Cl$_2$, respectively.

- Considering available IR and Raman spectra optical modes energies described the behaviour over the entire temperature range.

**Crystals at room temperature in a monoclinic space group $P_{2}^{1}/c$ with unit cells parameters $a = 8.213$ Å, $\beta = 93.72^\circ$, $c = 6.776$ Å.

- The structure at 123 K determined a low-temperature structure characterized by a space group $P2_{1}/c$ with the unit cell parameters $a = 8.272$ Å, $\beta = 91.07^\circ$, $c = 5.7516$ Å, $\gamma = 4$. The model describes the lattice sub system of a low dimensional magnet, which also has a low-dimensional structure (covalent chains/ladders connected by hydrogen bonds into a 3d structure).

**Monoclinic, space group $P2_{1}/c$ with the unit cell parameters $a = 7.323$ Å, $b = 12.725$ Å, $c = 9.768$ Å.

- The frequency is not function of wave vector k. It describes the contribution of optical modes to specific heat.

Motivation

Previous study of magneto-structural correlations of four CuIi based organo-metallic compounds with one-dimensional (1d) polymeric structure Cu(en)$_2$Cl$_2$, Cu(tn)$_2$Cl$_2$, Cu(en)$_2$SO$_4$, and Cu(en)(H$_2$O)$_2$SO$_4$ revealed that their magnetic subsystem has pronounced 2d character with effective intralayer coupling J$\approx 3$ K. The intra-layer spatial anisotropy of exchange coupling indicated in all compounds was described within the S = 1/2 Heisenberg model on the rectangular and zig-zag lattice with nearest-neighbor intra-layer couplings J$_{intra} < J$.

The purpose of this work is to study the lattice sub system of a low-dimensional magnet, which also has a low-dimensional structure (covalent chains/ladders connected by hydrogen bonds into a 3d structure).