Magneto-Structural Correlations and Field-Induced Phenomena in Cu(en)$_2$CrO$_4$

A. Darwich,[1] O. Vinnik,[1] R. Tarasenko,[1] E. Čižmár,[1] M. Orendáč,[1] and A. Orendáčová[1]

[1] *Institute of Physics, P. J. Šafárik University, Park Angelinum 9, 04001 Košice, Slovakia*

We report a magneto-structural study of the copper(II) coordination compound Cu(en)$_2$CrO$_4$ (en = C$_2$H$_8$N$_2$, ethylenediamine), which represents a system of weakly coupled spin-$\frac{1}{2}$ antiferromagnetic dimers. Structural analysis confirms the presence of a dimerized magnetic lattic in the system. Magnetic susceptibility, magnetization, and specific heat measurements were performed over a wide range of temperature and magnetic field to clarify the magnetic behaviour. Specific heat in zero magnetic field is well described by a simple Heisenberg model of isolated dimers with intradimer antiferromagnetic coupling $J/k_\mathrm{B} \approx -4.56$ K, with a spin gap of approximately 9.1 K, and shows no evidence of magnetic long-range order down to 0.4 K. The temperature-dependent single-crystal susceptibility of Cu(en)$_2$CrO$_4$ studied along the b axis shows a small deviation from the simple Heisenberg model of isolated dimers. Within a mean-field approach, the data are well described by an intradimer exchange coupling $J/k_\mathrm{B} \approx -4.56$ K, an interdimer interaction $z'J'/k_\mathrm{B} = -2.56$ K, and a g-factor $g_b = 2.15$. In the model of isolated dimers, the corresponding critical field closing the gap is about $B_c^b \approx 6.3$ T. Specific heat measurements in magnetic fields applied along the a axis uncover three distinct regimes, including the emergence of sharp λ-type anomalies above 6 T, indicative of a field-induced phase transition. These anomalies enabled the construction of a partial magnetic phase diagram, which is dome-shaped and symmetric relative to the center, Such character of the diagram is expected for unfrustrated interdimer interactions. Extrapolation of the phase diagram towards zero temperature yielded estimates of a critical field closing the gap $B_c^a \approx (5.5 \pm 0.5)$ T and a saturation field $B_\mathrm{sat}^a \approx (8.5 \pm 0.5)$ T. The experimental critical field is lower due to reduction of the spin gap by interdimer interactions ($g_a \approx g_b$). For further studies, first - principle calculations are necessary to determine the spatial distribution of interdimer interactions.