PRESSURE EFFECT ON CRYSTAL, MAGNETIC STRUCTURE AND VIBRATIONAL PROPERTIES OF VAN DER WAALS MATERIALS

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The discovery of long-range magnetic order in van der Waals materials down to the monolayer limit has established a new platform for the development of all-spintronic devices based on these structures and opened up new opportunities for studying physical properties of quasi-two-dimensional systems and low-dimensional magnetism, which holds significant fundamental and technological importance. The structural features of these compounds lead to a significant sensitivity of the physical properties to external influences, which can cause many unusual phenomena: charge, orbital and spin ordering, superconductivity, various phase transitions etc., which leads to special, exotic phenomena in them. In the last few years, Cr trihalides have been widely investigated, and a series of exciting phenomena are revealed in this family of materials. In this regard, further in-depth research and understanding of emerging properties in $CrBr_3$ and $CrCl_3$ becomes an essential objective, especially with variations in thermodynamic parameters.

This work focuses on the study of crystal, magnetic structure and vibrational properties of $CrBr_3$ and $CrCl_3$ in wide temperature and pressure ranges using neutron, X-ray powder diffraction and Raman spectroscopy. Neutron experiments were performed on the DN-6 diffractometer at the high-flux pulsed reactor IBR-2 (FLNP, JINR, Dubna, Russia).

In $CrCl_3$, a structural phase transition is observed under high pressure above ~2.3 GPa from the initial monoclinic $C2/m$ phase to the rhombohedral $R\bar{3}$ phase [1]. Anomalous behavior of some vibrational modes is also observed in the vicinity of the structural transition. Initial A-type AFM order in $CrCl_3$ is rapidly suppressed at high pressures of about 1 GPa. Instead, the FM state with a Curie temperature of $T_C \approx 19$ K is formed under high pressure. With further increase in pressure up to 2 GPa no evidence of magnetic contribution is observed.

Our results demonstrate that high pressure effectively drives a structural dimensionality crossover in $CrBr_3$ from a quasi-two-dimensional van der Waals system to a three-dimensional-like arrangement at $P \approx 15$ GPa [2]. Below this pressure, a strongly anisotropic compression is observed, with the interlayer distance decreasing rapidly while the layer thickness varies less markedly. Above 15 GPa, the compressibility becomes nearly isotropic and the interlayer distance reduces significantly more slowly, reflecting the onset of strong interlayer coupling. As a result, at high pressures the interlayer distance becomes smaller than the layer thickness, indicating a transformation from the weak interlayer coupling typical of van der Waals materials to a much denser three-dimensional bond. In addition, anomalies in the pressure dependence of several vibrational modes in the vicinity of the crossover were observed.

[1] O.N. Lis, D.P. Kozlenko, E.V. Lukin, S.E. Kichanov et. al. (2025) Pressure-induced ferromagnetism and structural phase transition in the van der Waals material $CrCl_3$, Phys. Rev. B. 112, 104105.

[2] D.P. Kozlenko, O.N. Lis, N.T. Dang, S.E. Kichanov, E.V. Lukin, et. al. (2025) Pressure induced crossover from 2D-like to 3D structural arrangement in van der Waals magnet $CrBr_3$, ChemPhysMater. 4, 280-288.