# Crystal and magnetic structure properties of van der Waals material $CrBr_3$ at extreme conditions

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## Relevance

The recent discoveries of magnetism in the monolayer limit have opened up new possibilities for the study of two-dimensional materials. Among layered transition-metal compounds, chromium tribromide, which crystal structure is comprising two-dimensional sheets of composition $CrBr_3$ van der Waals bonded to one another, is of particular interest due to its extraordinary electronic and magnetic properties, which is important for spintronic and magnetoelectronic applications etc.

Chromium trihalides have been known for many decades; however, they have received relatively little attention, especially of late, and are not particularly well understood. The crucial point is that $CrBr_3$ is a perfect model system to search for emergent physical phenomena, associated with the spin–lattice coupling in the $CrX_3$ family, also due to absence of structural phase transitions at low temperatures and similarity of magnetic order in bulk and few-layer forms.

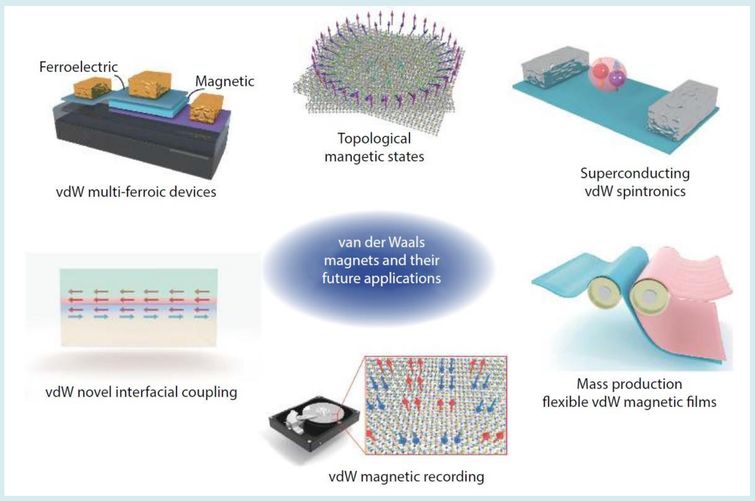
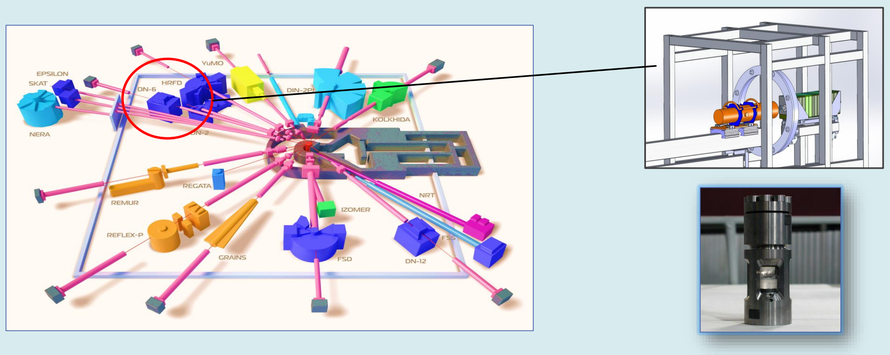



**Illustration of different nanostructures for vdW spintronics**

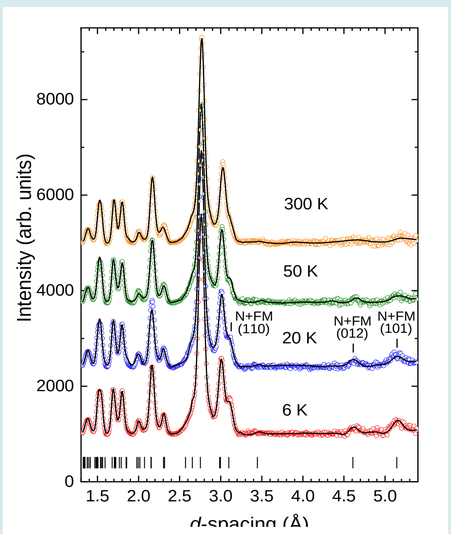
## Experiment

Detailed studies of the crystal structure of $CrBr_3$ were carried out using neutron diffraction on a DN-6 diffractometer of a pulsed high-flux IBR-2 reactor (FLNP, JINR, Dubna, Russia) in temperature range of 6-300 K and at pressure up to 5 GPa using the sapphire anvil high-pressure cell. The X-ray powder diffraction measurements were performed in the temperature range 15–300 K using the Malvern PANalytical Empyrean diffractometer with the Cu Kα radiation, λ = 1.541 Å

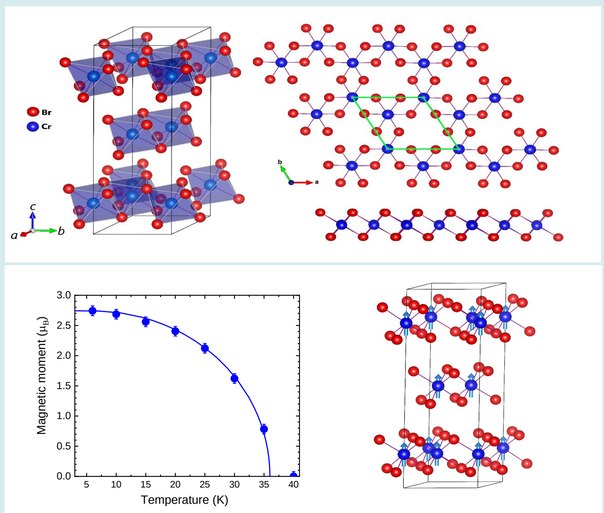



The Raman spectra with the single crystalline $CrBr_3$ samples were collected using a LabRAM HR Evolution spectrometer (Horiba, France) with a wavelength excitation of 632.8 nm emitted from He–Ne laser, 1800 grating. The low-temperature Raman measurements were carried out using low vibration helium refrigerator (Advanced Research Systems, USA) in temperature range 19–300 K.

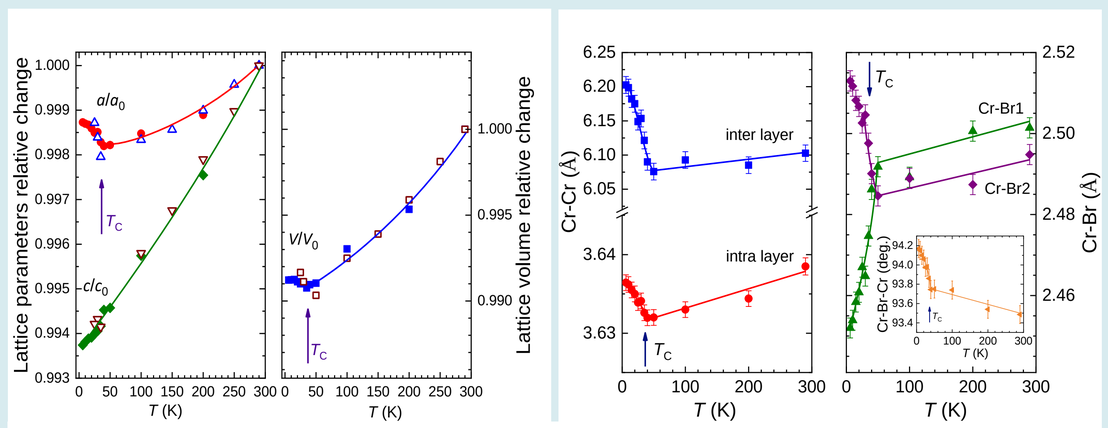

## Experimental results and discussion




Neutron diffraction patterns of $CrBr_3$ measured at selected temperatures

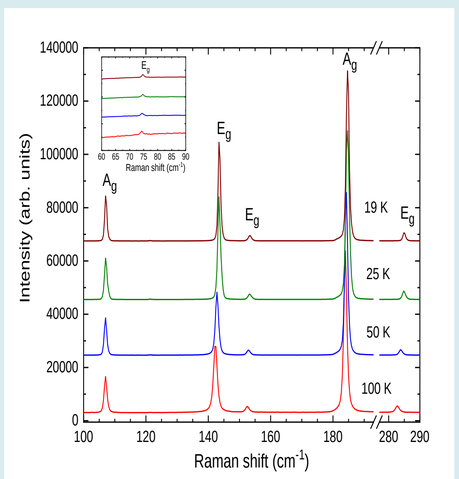



The rhombohedral structure of van der Waals crystal $CrBr_3$ of R3 symmetry, The $Cr^{3+}$-ordered magnetic moment as function of temperature and FM unit cell




a) The thermal expansion of $CrBr_3$ lattice is strongly anisotropic with the pronounced variation of the c lattice parameter. The temperature dependence of the a lattice parameter demonstrates anomalous behavior. Its thermal expansion changes from conventional positive character in the temperature range above $T_C$ to unexpected negative character below $T_C$.

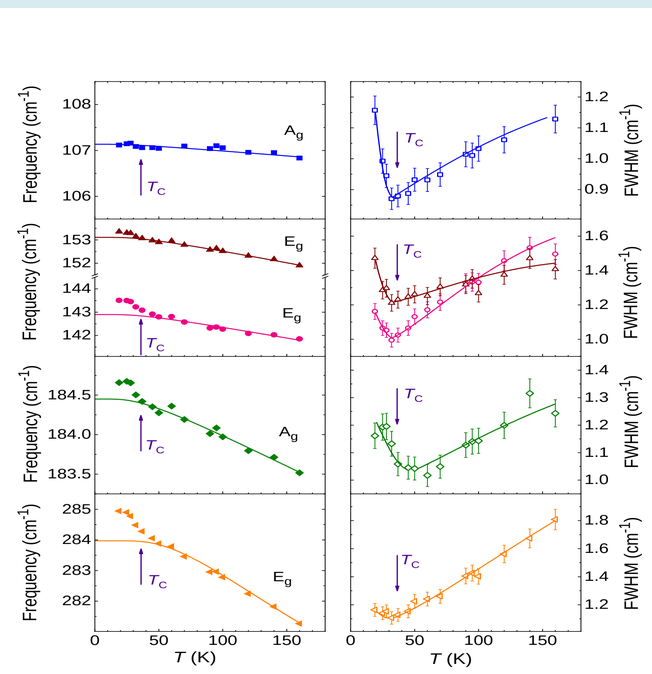
b) The interatomic intralayer and interlayer Cr–Cr distances decrease slightly on cooling in the temperature range above TC and they also demonstrate opposite increasing trend for $T < T_C$

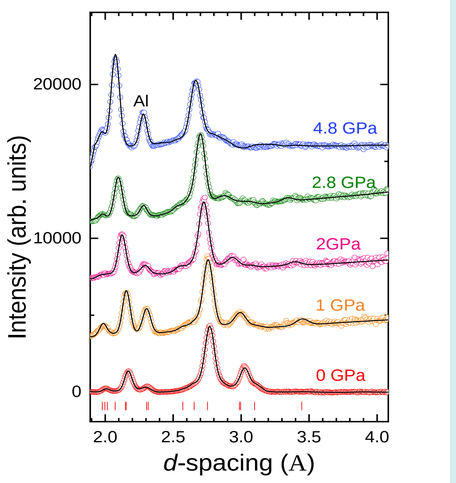



The Raman spectra, measured at selected temperatures

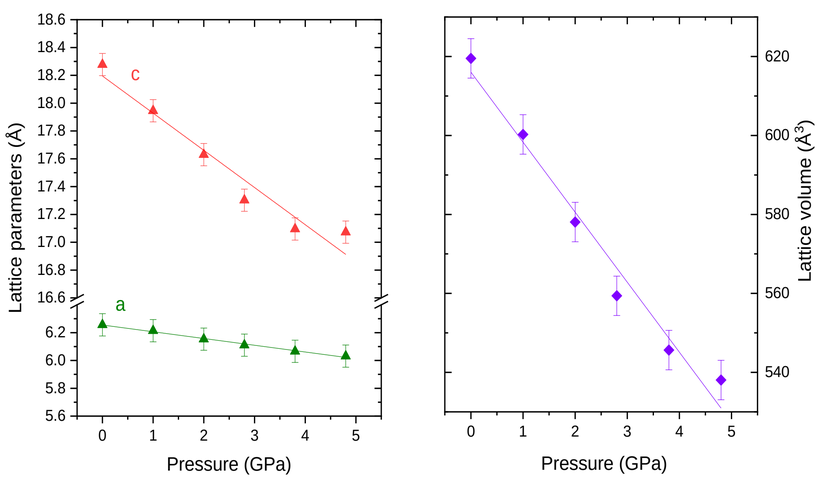
In the paramagnetic region, the frequencies of observed phonon modes demonstrate increasing trend with a temperature lowering, while full-width at half-maximum (FWHM) of relevant phonon peaks decreases.

The FWHM of all the observed phonon modes reach minimum in the vicinity of $T_C$ and demonstrate anomalous reversal broadening in the $T < T_C$ range. Both effects reveal a presence of the strong spin–phonon coupling in $CrBr_3$. The spin–phonon coupling is associated with the modification of the magnetic exchange interactions caused by the ionic motions, and the relevant




Neutron diffraction patterns of $CrBr_3$ measured at selected pressure




The baric dependences of the unit cell parameters of $CrBr_3$

The pressure dependencies of the unit cell parameters of $CrBr_3$ also show anisotropic behavior with the most compressible c parameter. At pressures above 3.8 GPa, there is an increase in peak intensity (116) at $d_{hkl}$ = 2.17 and a decrease of (110) at $d_{hkl}$ = 3.12, which may indicate some changes in the crystal structure, but it is difficult to determine from neutron data.