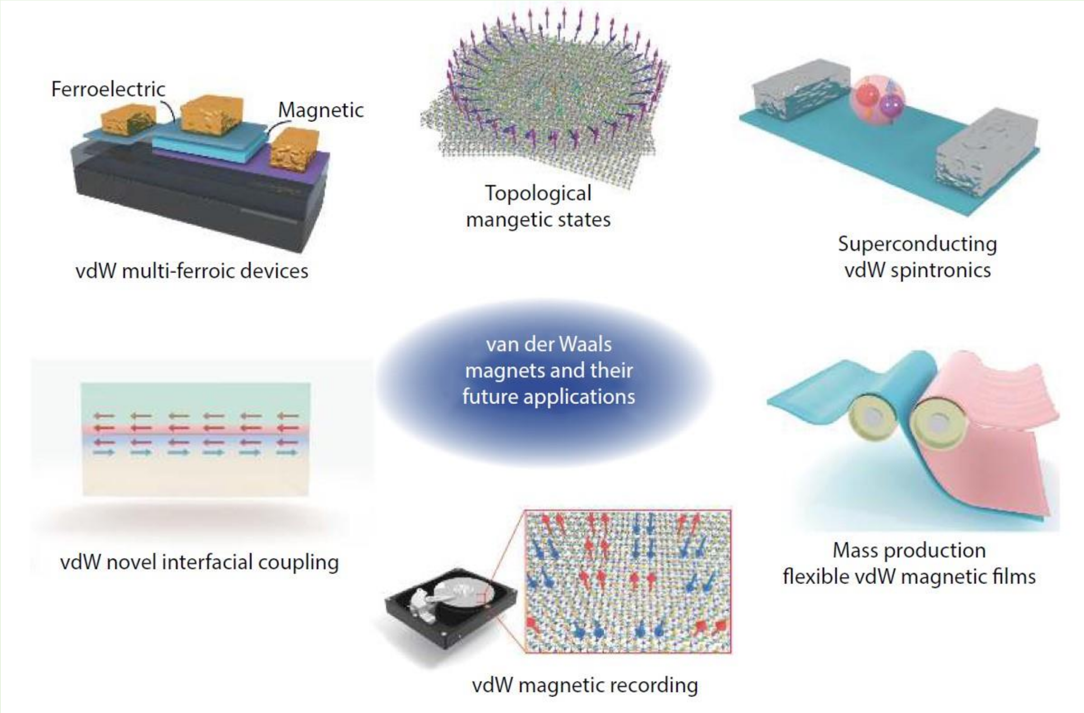
Crystal and magnetic structure properties of van der Waals material $CrBr_3$ at extreme conditions

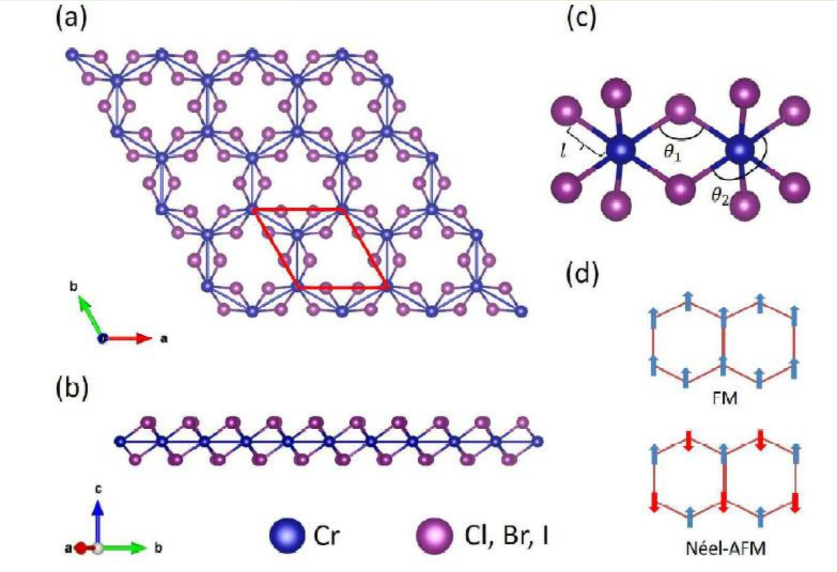
O. Lis, D.P. Kozlenko, S.E. Kichanov

Introduction

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.

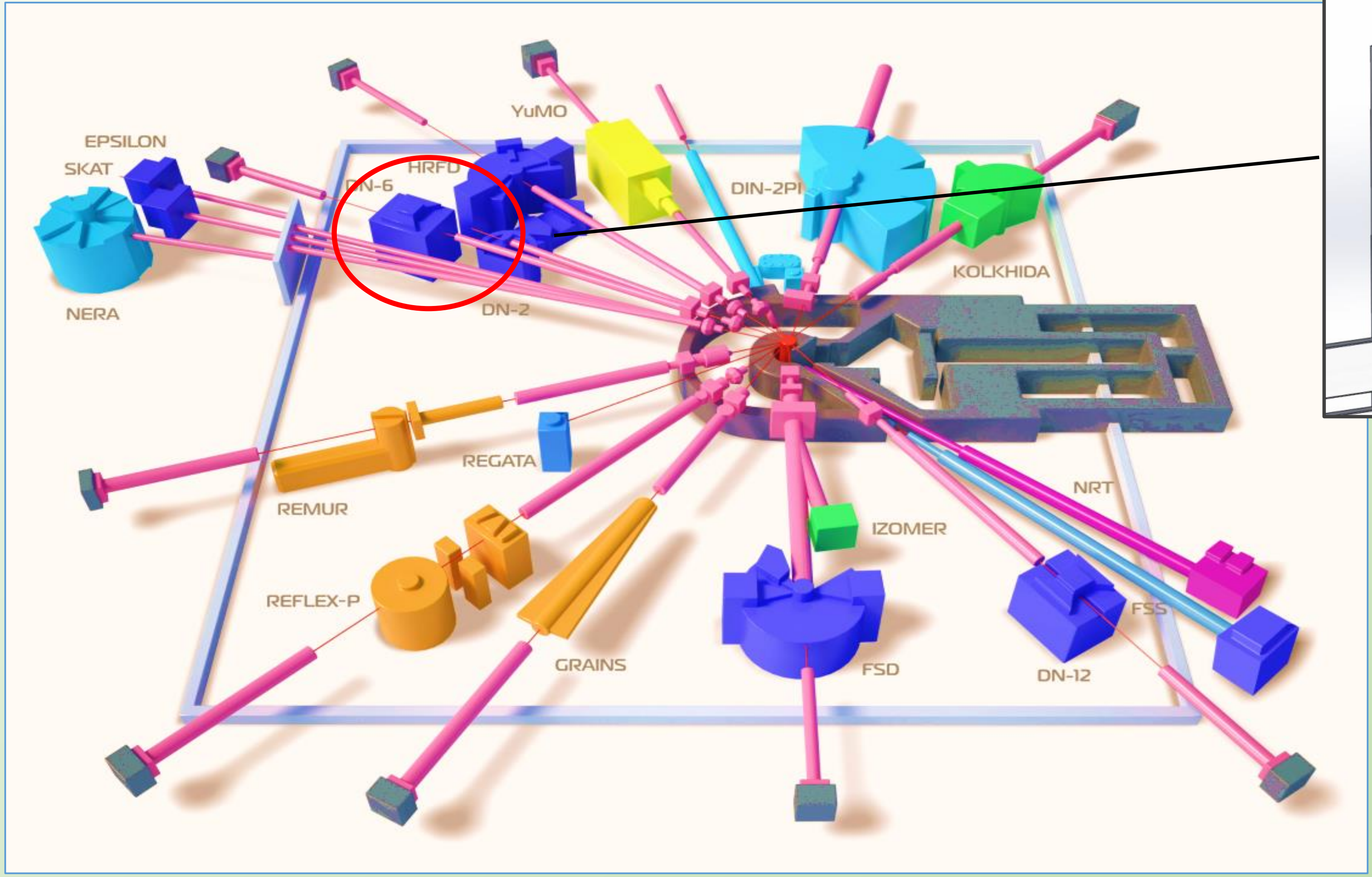
Chromium trihalides have been known for many decades; however, they have received relatively little attention, especially of late, and are not particularly well understood. The crucial point is that $CrBr_3$ is a perfect model system to search for emergent physical phenomena, associated with the spin–lattice 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.

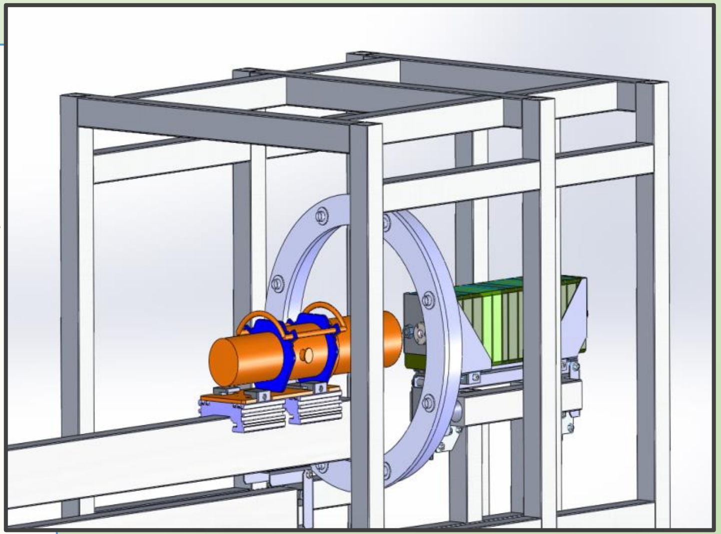




Atomic structure of monolayer CrX3. (a) Top view and (b) side view of a single layer; (c) Bonding between chromium and iodine atoms. The unit cell of CrI3 which includes two Cr and six I atoms has been indicated in (a). The bond length l between Cr and θ1 atom, the bond angle 1 between Cr and two I atoms in the same plane, and the axial angle θ2 are also shown in (c). The two magnetic orders, namely Neel-antiferromagnetic (AFM) and ferromagnetic (FM) are displayed in (d)

Experimental methods: Neutron diffractometer DN-6





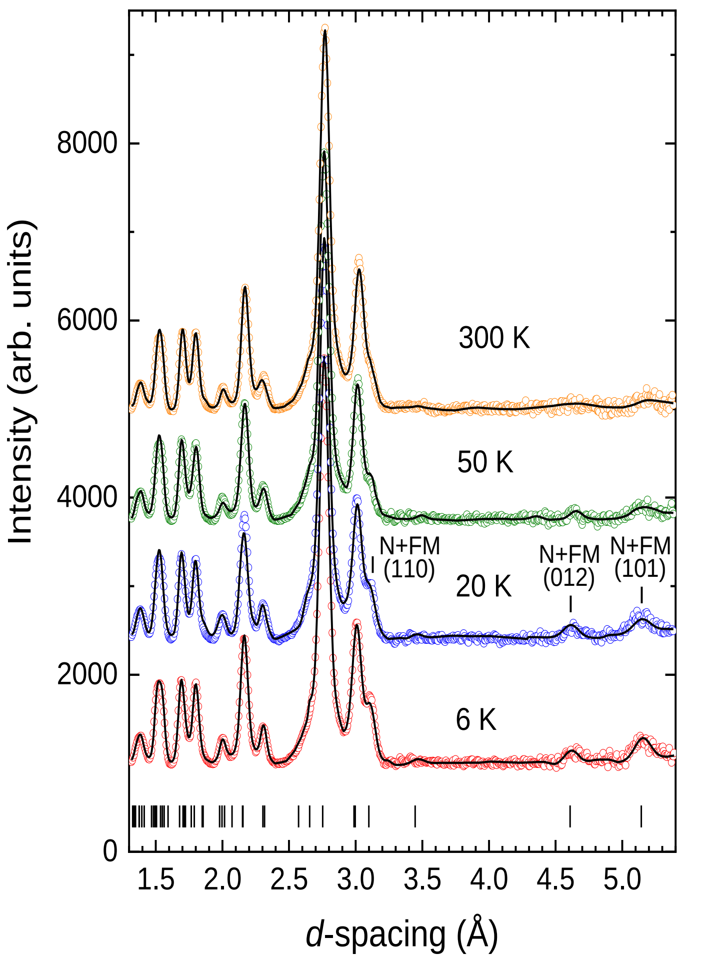
Experimental hall of the IBR-2 reactor with 14 neutron output channels and the layout of the DN-6 diffractometer

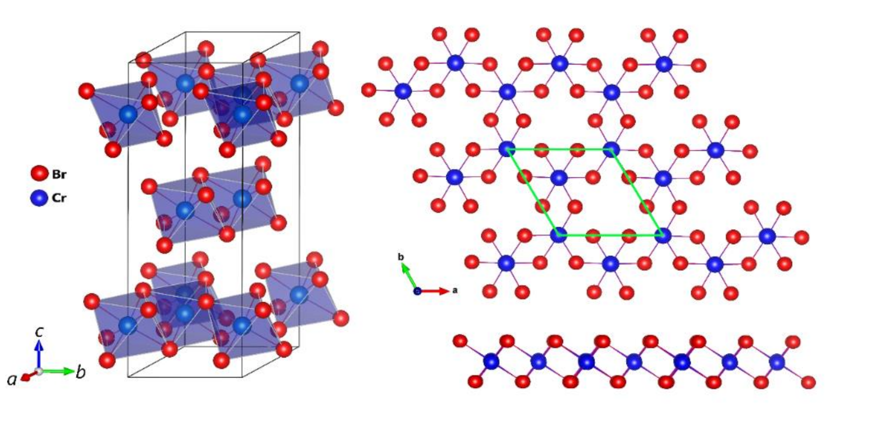
High-pressure cell with sapphire anvils

Experimental methods: Raman spectroscopy

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.

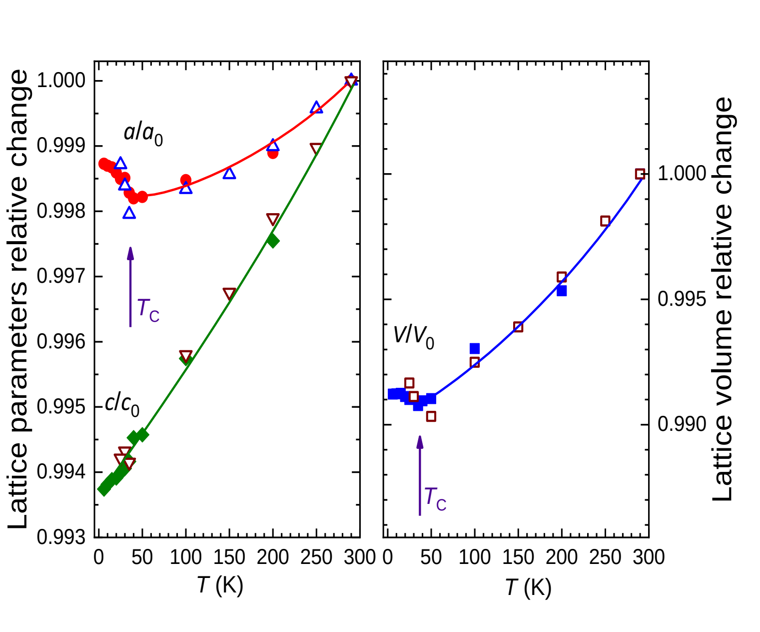
Neutron diffraction at low temperature

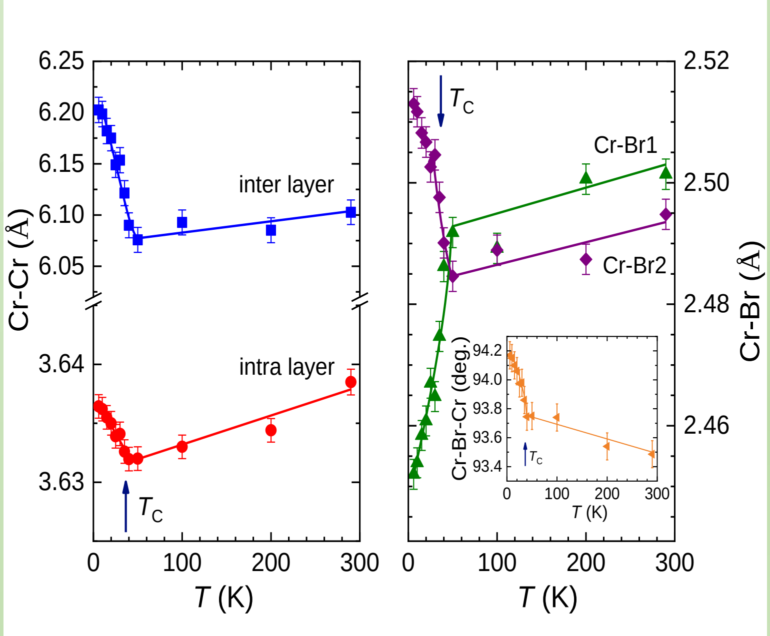




a) The thermal expansion of $CrBr_3$ lattice is strongly anisotropic with the pronounced variation of the c lattice parameter.

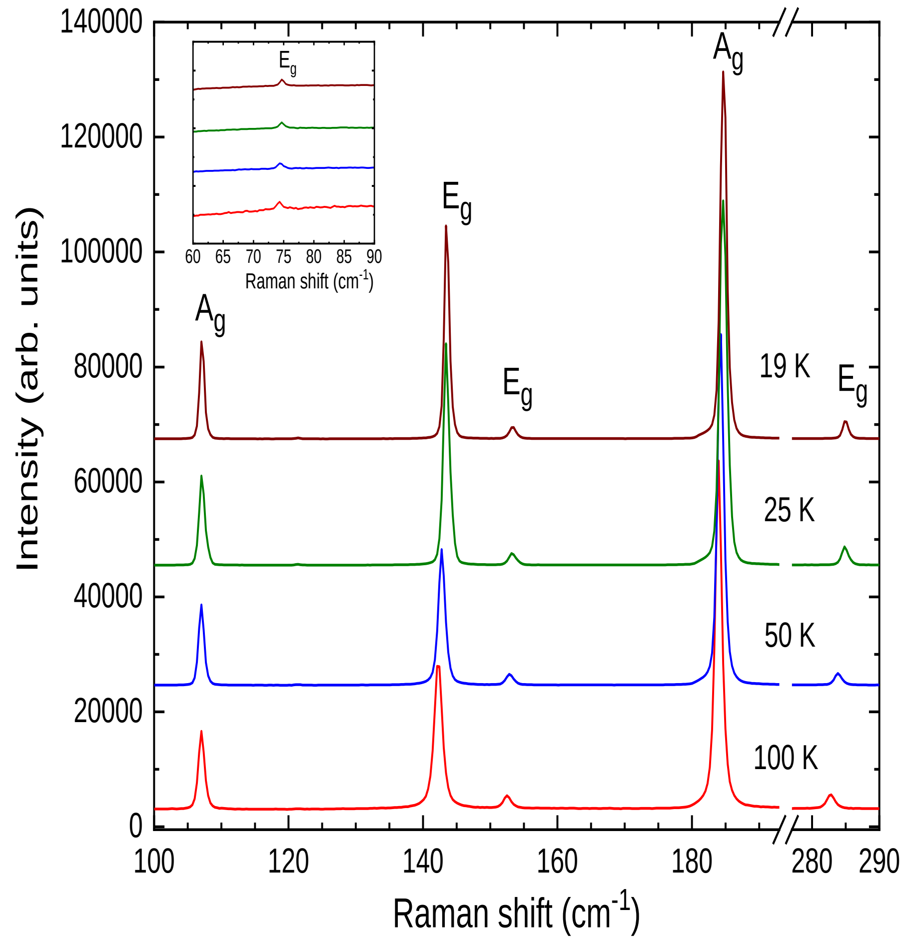
b) The interatomic intralayer and interlayer Cr–Cr distances decrease slightly on cooling in the temperature range above TC and they also demonstrate opposite increasing trend for T < T_C

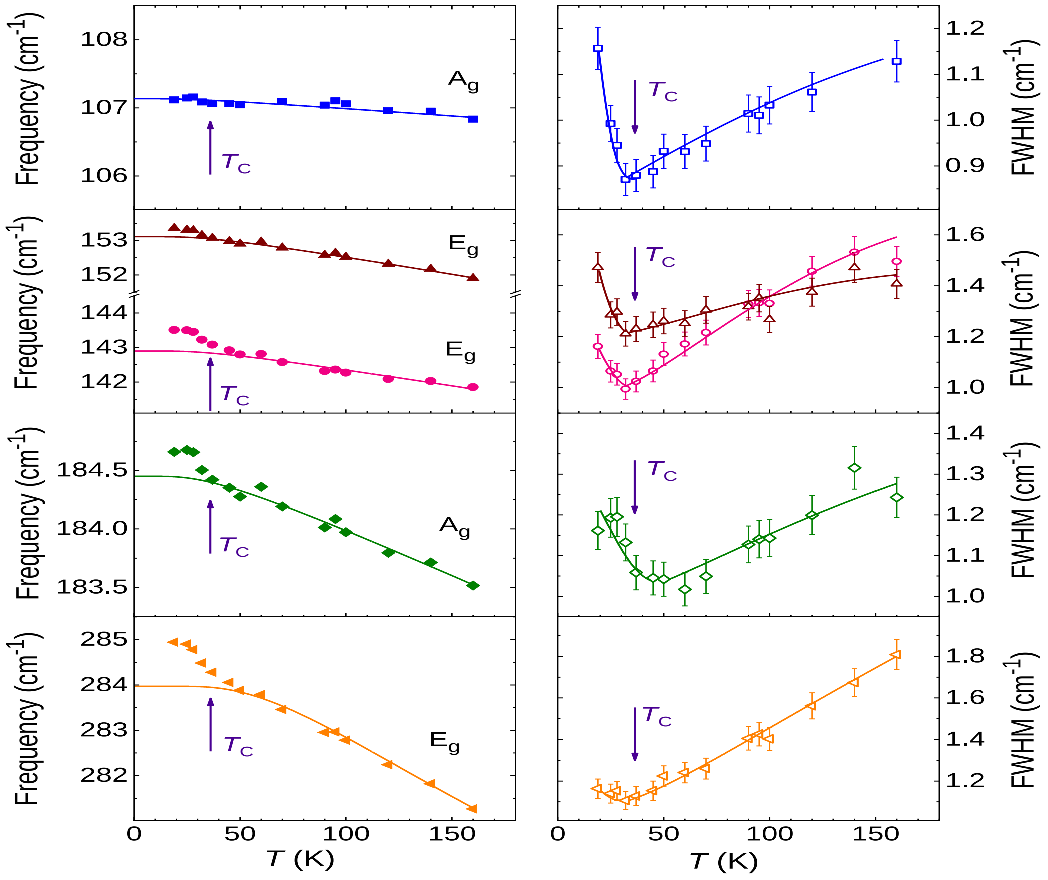




Kozlenko, D.P., Lis, O.N., Kichanov, S.E. et al. npj Quantum Mater. 6, 19 (2021)

Raman spectroscopy at low temperature

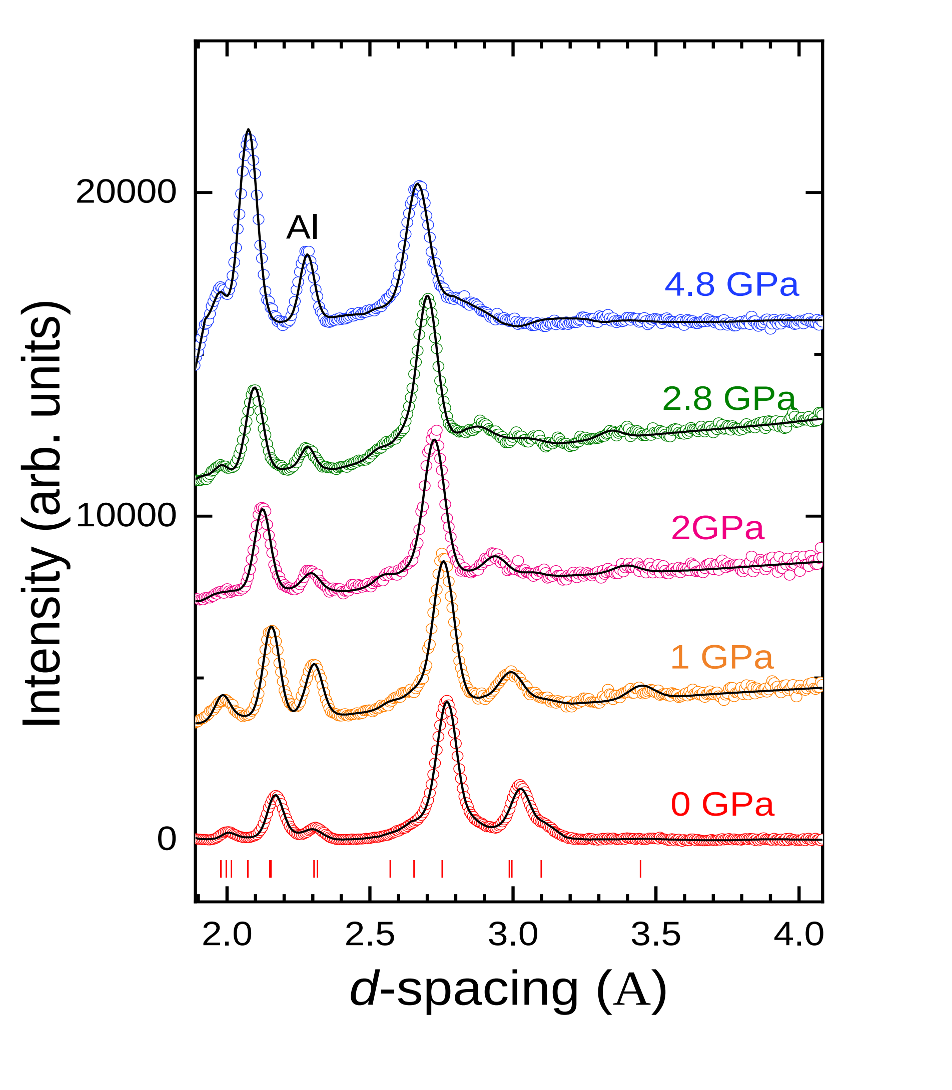




In the paramagnetic region, the frequencies of observed phonon modes demonstrate increasing trend with a temperature lowering, while full-width at half-maximum (FWHM) of relevant phonon peaks decreases.

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 at high pressure



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