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Raman Spectroscopy Study of magnetic Quasi-two-dimensional materials

Quasi-two-dimensional materials, known for their easy exfoliation to a monolayer and unique optical and transport properties are promising candidates for nanospintronics and nanoelectronics. Experimental confirmation of magnetic ordering persisting to a monolayer in these complex systems in 2017 did not only widen up the area of their potential application, but also opened up a completely new experimental field in condensed matter physics. Considering that previously widely accepted theories forbid magnetic ordering in 2D systems, it is no surprise that magnetic quasi-2D materials have only recently become an area of extensive study. Aiming to provide much needed insight into these systems, we have performed Raman spectroscopy studies of CrI₃ and VI₃ single crystals. Polarization dependent Raman spectra of CrI₃ single crystals were analyzed in accordance with suggested low-temperature and high-temperature structures, confirming the existence of phase transition between the low-temperature rhombohedral and the high-temperature monoclinic structure [1]. In the temperature dependence of phonon energies and line widths a clear splitting of the E_g rhombohedral modes into the A_g and B_g monoclinic modes can be observed at 180 K, contrary to the previously reported 220 K. No phase co-existence can be tracked within our spectra. Polarization dependent Raman spectra, together with DFT calculations and PDF analysis of synchrotron XRD patterns, provided an answer to the on-going debate regarding the crystal structure of VI₃ [2]. Our results point to two possible scenarios: the coexistence of two phases, short range ordered P3̄1c and long range ordered R3̄ as two segregated phases and/or randomly distributed short range ordered P3̄1c domains in the long-range ordered R3̄ lattice. Observed phonon line asymmetry of the most pronounced peak indicates strong spin-phonon coupling in this quasi-two-dimensional material.

References

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