Phase transitions in ferroelectric van der Waals crystals under pressure

Nada Alghofaili [1,2], Anna Kusmartseva [3], Mark T. Greenaway [3] and Amalia Patané[1]

1) School of Physics & Astronomy, University of Nottingham, Nottingham NG7 2RD, UK
2) Department of Physics, College of Sciences, Qassim University, Buraydah 52571, Saudi Arabia
3) Department of Physics, Loughborough University, Loughborough, LE11 3TU, UK

Van der Waals (vdW) crystals consist of weakly interacting atomically thin layers bound by weak vdW forces. They can be stacked and twisted in different ways to create heterostructures that can exhibit a wide range of different phenomena, including emergent properties such as interfacial ferroelectricity [1]. VdW crystals, such as α-In_2Se_3, can also be intrinsically ferroelectric. α-In_2Se_3 is a semiconductor with non-centrosymmetric vdW layers that exhibits both out-of-plane and in-plane ferroelectric polarization at room temperature [2]. Furthermore, it is reported to have multi-ferroic order combining ferromagnetism, ferroelectricity, and ferroelasticity [3]. The different polymorphs and polytypes phases of In_2Se_3 feature distinct structural and ferroelectric properties [4], which are known to be affected by pressure [5]. Here, we report on structural phase transitions in α-In_2Se_3 due to hydrostatic compression and decompression [6]. We probe the behaviour of the phase transition by in situ Raman spectroscopy and imaging in a high-pressure diamond anvil cell (Figure 1). Mechanical deformations of the crystal lattice and their layer stacks (2H or 3R with hexagonal ($P6_3mc$) and rhombohedral (R3m) space group symmetry, respectively) are modelled by density functional theory, revealing a phase transition into the paraelectric b-phase of In_2Se_3. Our investigation reveals that the behaviour of 2H-α-In_2Se_3 is significantly different to that of 3R-α-In_2Se_3 [5,6]. Specifically, we find that the reversibility of the ferroelectric-paraelectric phase transition depends on the sample quality and the underlying lattice strain induced by defects and imperfections. Understanding and controlling the coupling between the electrical and mechanical properties of ferroelectric vdW crystals is crucial for the development of technological applications, spanning from sensors and actuators, to memory and fast switching devices.

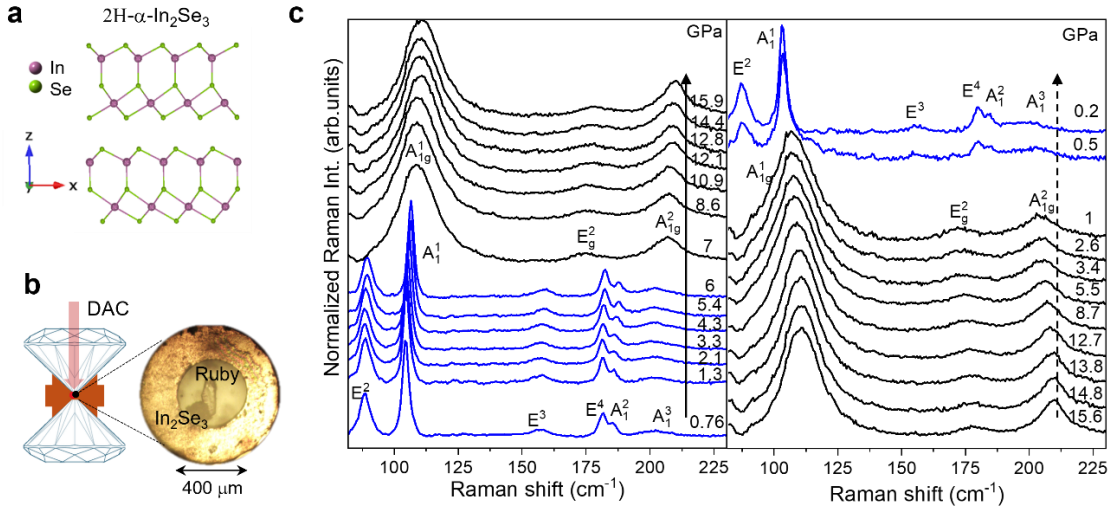

Figure 1: a) Crystal structure of 2H-α-In_2Se_3, b) Raman spectroscopy inside a DAC, c) Raman mode evolution under pressure.

[1] Weston A. et al., Nat. Nanotechnol. 17 (2022) 390
[2] Ding W. et al., Nat. Commun. 8 (2017) 1499
[3] Liu C. et al., Appl. Phys. Lett. 118 (2021) 072902
[4] Wu Y. et al., Nat. Commun. 15 (2024) 10481
[5] Vilaplana R. et al., Inorg. Chem. 57 (2018) 8241–8252
[6] Nada Alghofaili et al., 2D Mater. 13 (2026) 025025