Phonon Signatures of Instabilities in van der Waals material $InSiTe_3$

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Although van der Waals materials are widely studied for their unique properties and applications, $InSiTe_3$ remains relatively neglected despite being synthesized over 30 years ago, especially when compared to the extensive research on related compounds like $CrSiTe_3$ and $CrGeTe_3$. Here, we present a detailed experimental and theoretical investigation of this van der Waals material. Single-crystal X-ray diffraction reveals the rhombohedral crystal symmetry of InSiTe3, described with the $P\bar{3}$ space group. Polarization-dependent inelastic light scattering experiments further validate the symmetry and reveal pronounced anharmonicity in $InSiTe_3$. Namely, in addition to the symmetry predicted Raman active modes, which show excellent agreement with DFT calculations, the spectra exhibit unexpected features; tripling of a localized high-energy A_g mode at approximately 500 cm^{-1}. For E_g modes the extracted phonon-phonon coupling constants indicate moderate anharmonicity. However, for A_g modes, the coupling strength increases dramatically, reaching values up to ten times higher than those of the doubly degenerate modes. The intensities of the analysed A_g modes display anomalous behaviour with increasing the temperature, particularly around 200 K. Notably, at this same temperature, higher-order overtone excitations emerge within the gap of the calculated phonon density of states. Having in mind that phonons couple to various degrees of freedom, these interactions leave a distinct fingerprint on their behaviour. While our findings highlight these instabilities, its origin lies beyond the scope of this research.

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