Absence of charge density wave transition in $SnSe_2$ at extreme conditions

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Abstract

Transition Metal Dichalcogenides (TMDs) have attracted significant attention as their layered crystal structure results in anisotropic mechanical and electronic properties and into the quantum confinement of charges into single layers, making them prototypical two-dimensional (2D) materials [1, 2]. In recent years, high-pressure (HP) studies on TMDs showed that the pressure-induced enhancement of the weak inter-layer interaction among adjacent layers can strongly modify their structural and electronic properties [3-6], resulting in a rich phase diagram which can host a metallic transition in semiconducting TMDs [3- 5] or more exotic phenomena at high-pressure/low-temperature (LT) conditions, such as charge density waves (CDWs) or superconducting states [3, 6]. Among them, $SnSe_2$ has recently gained attention due to the unusual CDW-like phenomenology emerging during the metallization process, consisting of a periodic lattice distortion (PLD) witnessed by X-ray diffraction [6] and the high-pressure reorganisation of the band structure leading to the suppression of large parts of the Fermi surface suggested by Density Functional Theory calculations [7].

In our work, we seek experimental evidence of a CDW-like ordering by investigating the evolution of the inter-band electronic transitions, the charge carriers, and the vibrational spectrum of $SnSe_2$ by synchrotron-based infrared transmission and reflectivity high-pressure measurements and by LT/HP Raman spectroscopy. By IR spectroscopy, we demonstrate a clear metallization above 16 GPa, with the closure of the bandgap and the subsequent increase in the Drude plasma frequency in the far-IR region. The metallization is accompanied by a change in the Raman spectrum ~16 GPa, in accordance with the literature [6]. On increasing pressure up to 27 GPa, we find no evidence for the presence of CDW neither in the IR spectra, displaying no signatures of Fermi-level gapping, nor in the LT/HP Raman measurements, lacking CDW-related spectral contributions, such as the amplitude mode.

Our IR and Raman spectroscopic studies of $SnSe_2$ provide an exhaustive picture of the electronic, optical and vibrational evolution of $SnSe_2$ under extreme pressure and temperature conditions, without finding clear evidence support for the formation of an unusual pressure-induced CDW in the investigated 0-27 GPa and 300-65 K ranges.

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