## **Suplementary Pages (Optional)**

Surface phonon polaritons (SPhPs) are electromagnetic surface waves that emerge when photons couple with optical phonons in a polar dielectric material<sup>1</sup>. SPhPs are mid- to farinfrared (IR) analogs of surface plasmon polaritons (SPPs) which emerge when photons couple with the free carrier plasma in a metal. Although SPPs and SPhPs are both resonant surface modes which can remarkably enhance the near-field electromagnetic density<sup>2, 3</sup>, SPhPs suffer from lower losses due to the relatively long-lifetime of optical phonons.4–6 The ability of SPhPs to mediate light-matter interactions is usually manipulated by nanostructuring the material interface in order to modify the near-field electromagnetic density of states (EMDOS). Nanostructuring polar dielectrics to design SPhP modes is thus a promising approach to engineer near-field optical phenomena. In particular, SPhPs enable the confinement of electromagnetic fields below the Abbe diffraction limit, providing a way to realize high quality-factor resonances. Sub-diffraction light confinement has already facilitated the development of the field of nanophotonics and promises further advances in applications such as surface-enhanced spectroscopy techniques (such as SEIRA and SERS), free-space IR communications, and superlensing<sup>4</sup>.

In this work, we demonstrate 3D Raman mapping of surface electromagnetic modes in InP nanopillars and 1D gratings of 4H-SiC, using a standard confocal Raman microscopy setup. We further demonstrate that the polarizability associated with SPhP modes investigated is consistent with the Raman polarizability selection rules. Finally, we illustrate the ability of Raman mapping to investigate electron-phonon coupling via the longitudinal optical plasmon coupled (LOPC) effect.



Supplementary Figure 1: Top Panel: (Left) Theoretical and (right) experimental maps of the LO and TO phonon modes for a xy cross-sectional view. The edge of the nanopillar is denoted by dotted green lines.

Bottom Panel: Same as top for a yz cross-sectional view.