## Multidirection Piezoelectricity in Mono- and Multilayered Hexagonal a-In2Se3

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Piezoelectric materials have been widely used for sensors, actuators, electronics, and energy conversion. Two-dimensional (2D) ultrathin semiconductors, such as monolayer h-BN and MoS2 with their atom-level geometry, are currently emerging as new and attractive members of the piezoelectric family. However. their piezoelectric polarization is commonly limited to the inplane direction of odd-number ultrathin layers, largely restricting their application in integrated nanoelectromechanical systems. theoretical calculations have Recently, predicted the existence of out-of-plane and in-plane piezoelectricity in monolayer a-In2Se3. Here, we experimentally report the coexistence of out-of-plane and in-plane piezoelectricity in monolayer to bulk a-In2Se3. attributed to their noncentrosymmetry originating from the hexagonal stacking. Specifically, the corresponding d33 piezoelectric coefficient of a-In2Se3 increases from 0.34 pm/V (monolayer) to 5.6 pm/V (bulk) without any odd-even effect. In addition, we also demonstrate a type of a-In2Se3-based flexible piezoelectric nanogenerator as an energy-harvesting cell and electronic skin. The out-of-plane and in-plane piezoelectricity in a-In2Se3 flakes offers an opportunity to enable both directional and nondirectional piezoelectric devices to be applicable for self-powered systems and adaptive and straintunable electronics/optoelectronics.