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Ultrafast Electrochemical Synthesis of Defect-Free In_2Se_3 Flakes for Large-Area Optoelectronics

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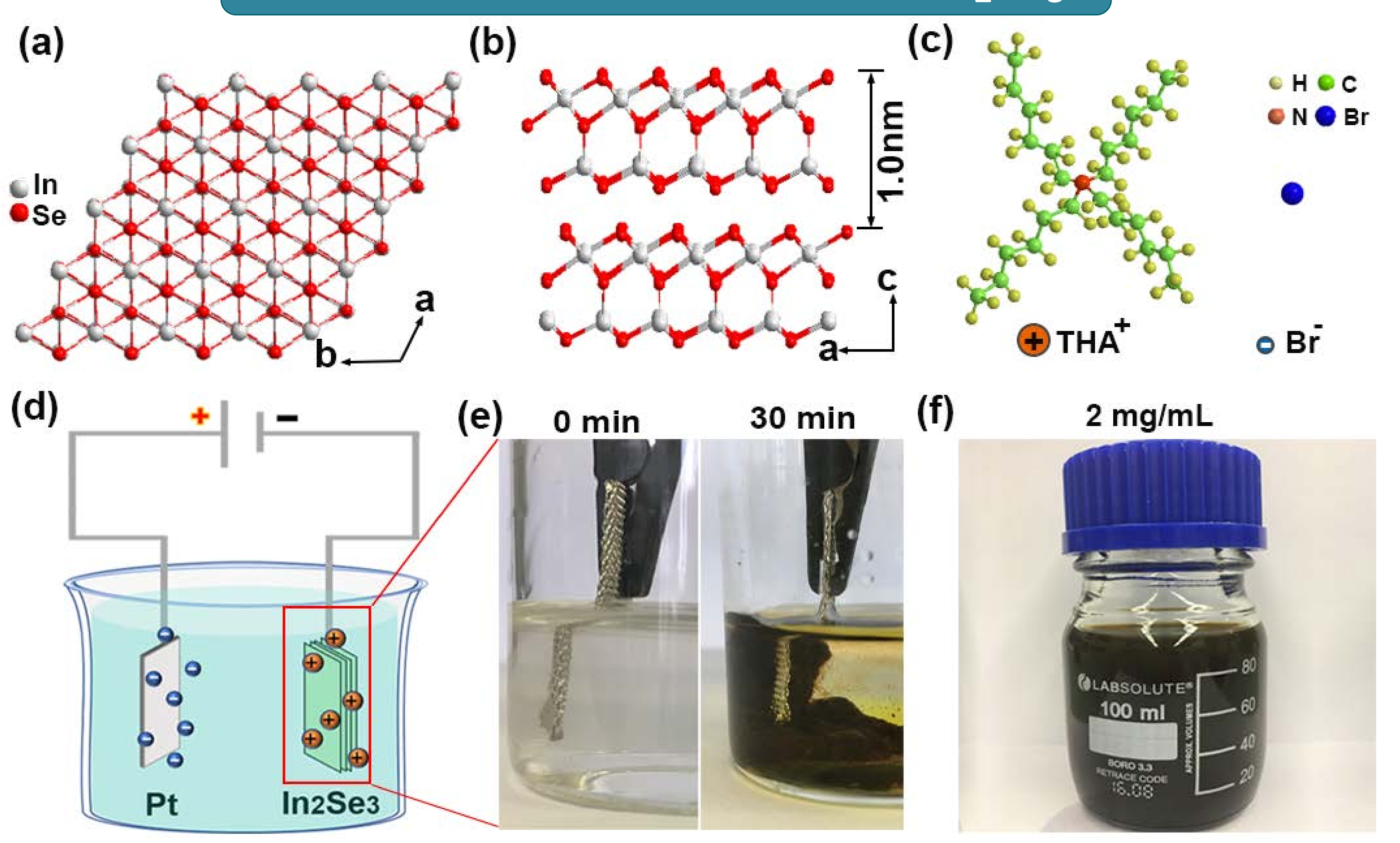
Introduction

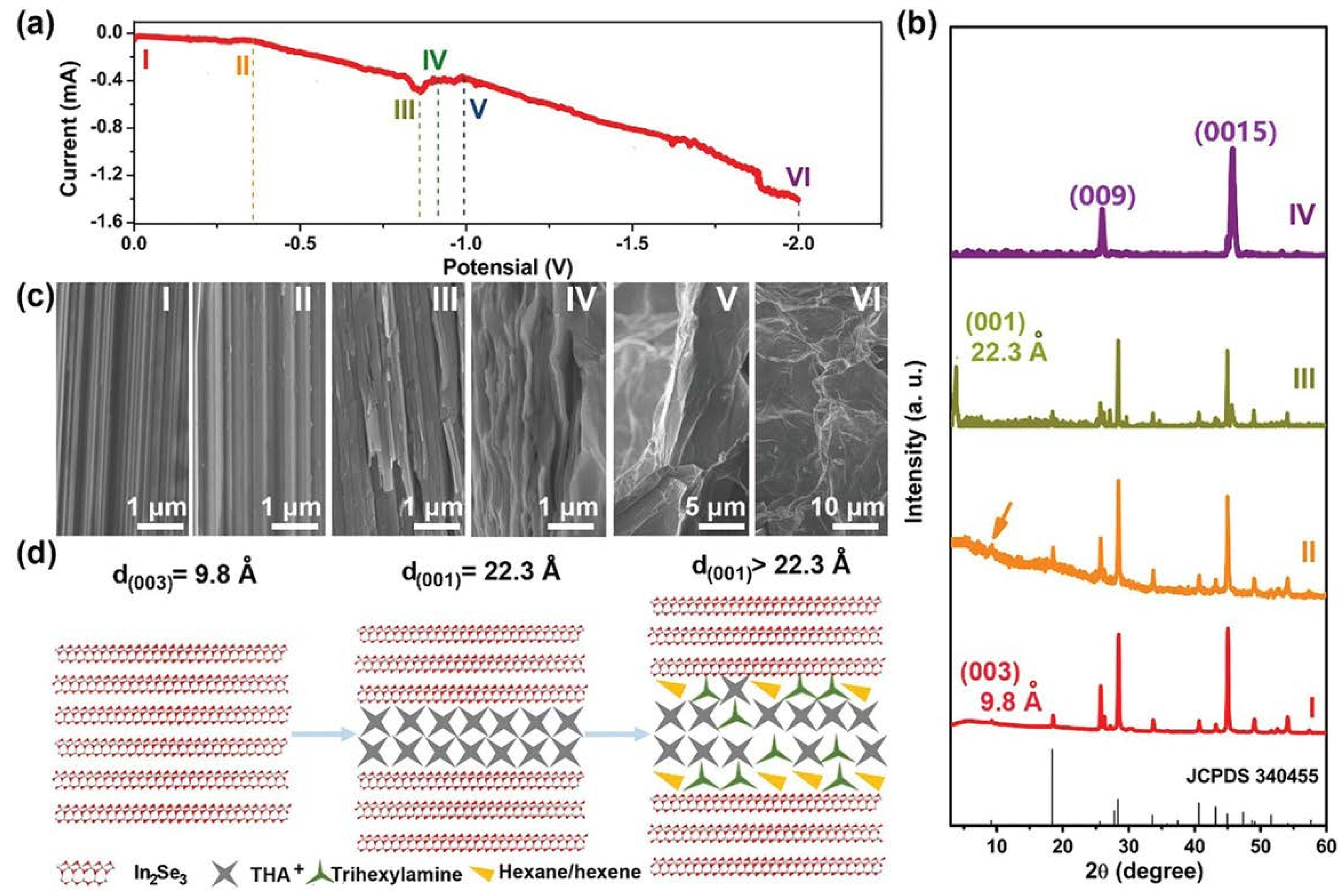
Indium(III) selenide (In_2Se_3), an important semiconductor, has been applied in various electronics and optoelectronics owing to its thickness-dependent direct bandgap (1.3-1.7eV) and exceptional optoelectronic properties and high stability[1]. However, the scalable production of defect-free In_2Se_3 flakes with large crystal domains remains an impediment to their practical applications. Here, a facile electrochemical strategy is presented for the ultrafast delamination (30 min) of bulk layered In_2Se_3 crystals in dimethylformamide (DMF) containing (tetrahexylammonium(THA$^+$), resulting in high-yield (83%) production of In_2Se_3 flakes with large lateral size (up to 26 µm). The intercalation of THA$^+$ ions mainly creates stage-3 intercalated compounds in which every three layers of In_2Se_3 are occupied by one layer of THA molecules[2]. The subsequent exfoliation leads to a majority of trilayer In_2Se_3 nanosheets (4 nm). Owing to the excellent solution processability of exfoliated sheets (2 mg/mL in DMF), large-area (400 µm × 20 µm) photodetectors are fabricated based on filtrated In_2Se_3 thin films from their stable dispersions. The fabricated devices demonstrated high responsivity (≈1 mA W^{-1}), superfast rise (41 ms) and decay time (39 ms) and high stability to light, superior to the performances of other 2D materials such as, graphene, black phosphorus, MoS_2, and WS_2[3].

Electrochemical exfoliation of In_2Se_3



1. Cathodic electrochemical intercalation: THA$^+$ in DMF
2. Ultrafast intercalation: 30 mins
3. High concentration (2 mg/mL) of In_2Se_3 dispersion in different solvents (like, DMF, NMF, IPA and Acetonitrile) 4. High yield: 83 %

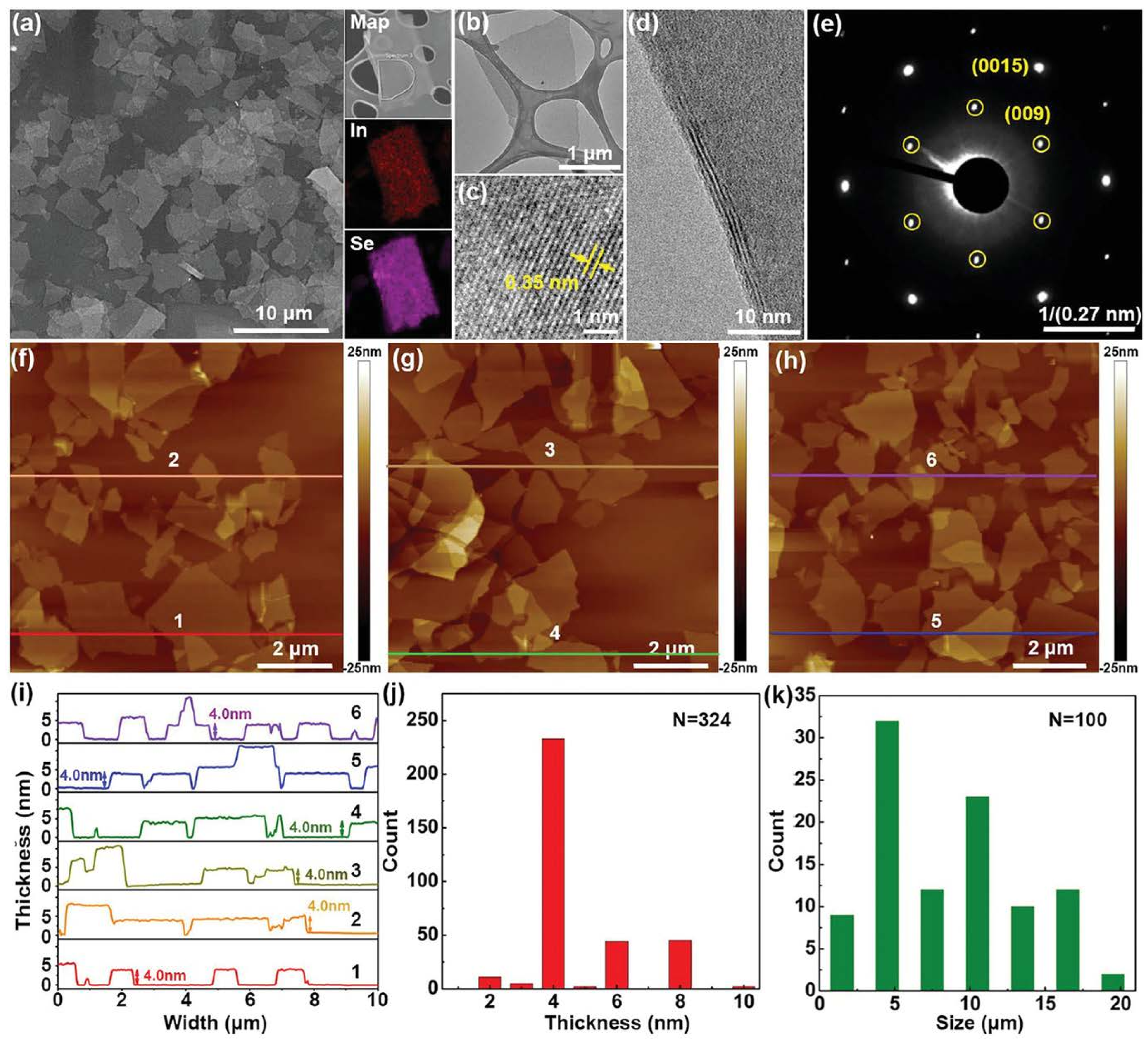
Mechanism of intercalation process



$In_2Se_3 + xTHA^+ + xe^- \rightarrow (THA^+)_x In_2Se_3^{x-}$

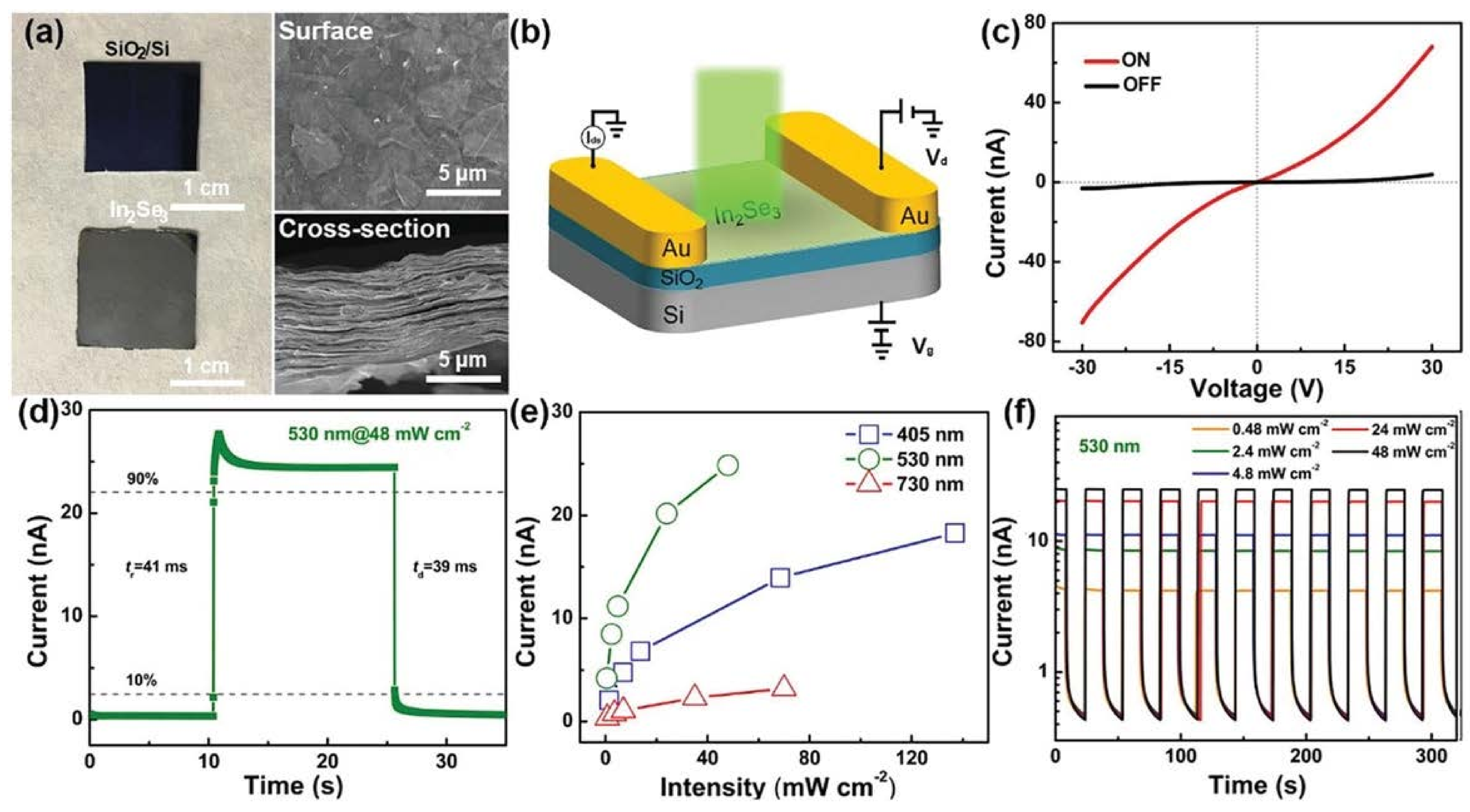
$(THA^+)_x In_2Se_3^{x-} \rightarrow In_2Se_3$+trihexylamine+alkanes (e.g., hexane)

Structure characterization of In_2Se_3 flakes



In_2Se_3 flakes: average size: 8.6 µm; average thickness 4.0 nm (3 layers); defect-free flakes confirmed by HR-TEM

Thin-film photodetectors based on In_2Se_3



1. The size of device: 400 µm × 20 µm
2. Efficient responsivity (1 mA W^{-1}) and ultrafast response time with a rise and decay of 41 and 39 ms.
3. Great potential for the future development of photonic devices

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