

Supplementary Information

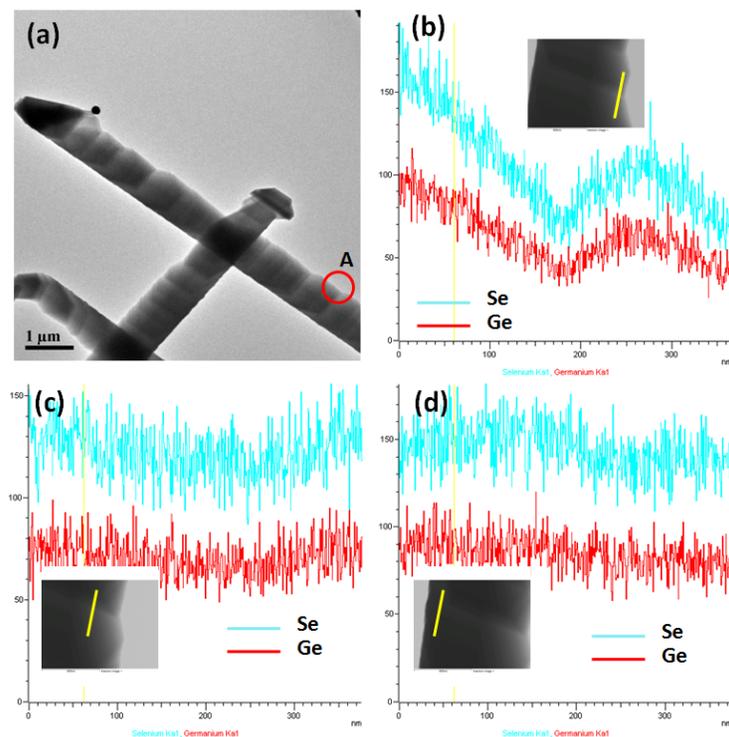


Figure S1. (a) Low-magnification TEM image of three GeSe₂ NBs; (b-d) The EDS composition profiles at different axial positions of the stepped-surfaced NB of location A (as indicated with red circled line) in Fig. S1 (a) demonstrates a uniform distribution of the compositional elements Ge and Se with atomic ratio 1:2 stoichiometry. The insets show the low magnification TEM images of the NB with the labeled lines along which the EDS composition profiles were taken.

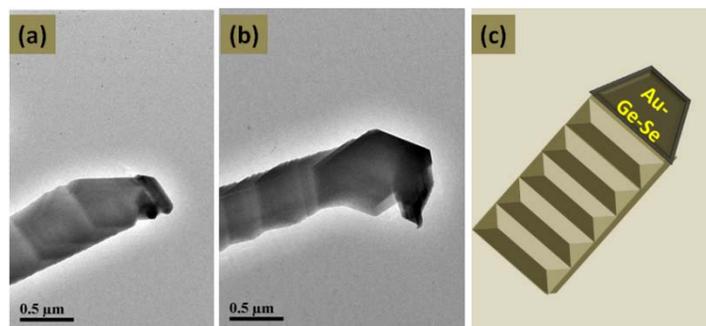


Figure S2. (a, b) The low magnification TEM images of different shaped catalyst particles formed on the tips of the NBs. (c) The schematic diagram of the single NB with the stepped surfaced along the length.

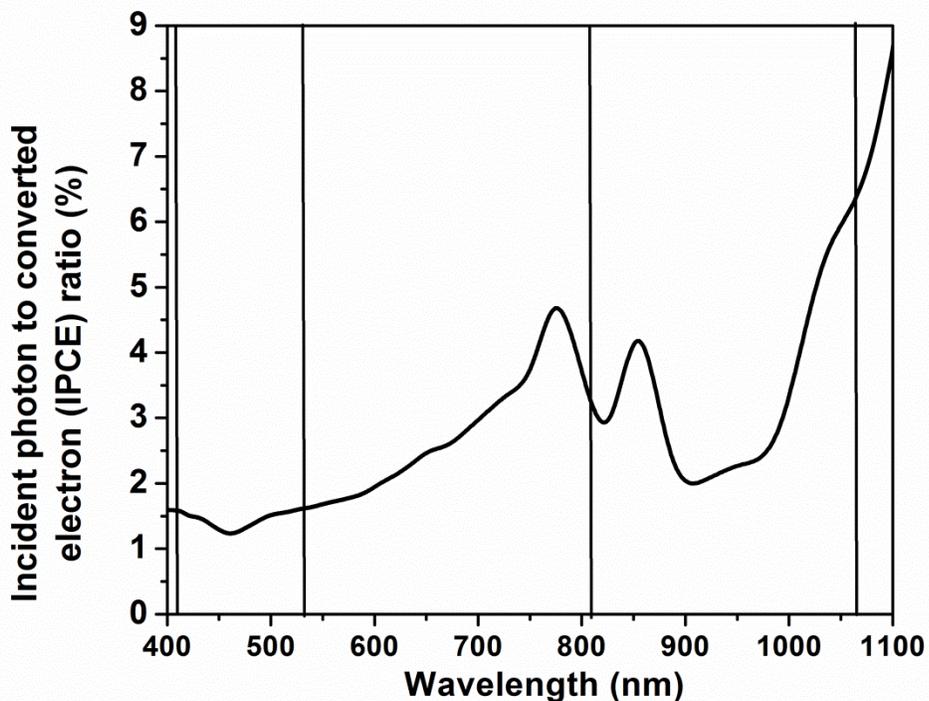


Figure S3. Measured IPCE spectra of individual stepped surfaced GeSe₂ NB device at the incident wavelength range 400 to 1110 nm at a fixed zero bias.

To quantitatively address the photoactivity of individual GeSe₂ NB device, we performed incident-photon-to-current-conversion efficiency (IPCE) measurements to study the device photoresponse as a function of incident light wavelength (**Figure S3**). IPCE is expressed as $IPCE = (1240J)/(J_{light}\lambda)$, where J is the photocurrent density from the device at zero external bias, λ is the incident light wavelength, and J_{light} is the incident light power density. The IPCE was measured by an IPCE measurement kit (Newport –Model 77890). The NB device shows lower IPCE value at visible wavelength range (400-500 nm) and relatively higher IPCE value in IR wavelength range, which is well agreed with our observation from discrete laser sources photocurrent studies.