Supporting Information

S1. Structural determination of CdSe nanobelts by X-ray diffraction (XRD)

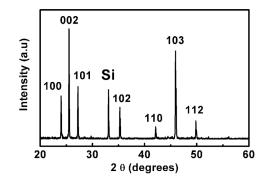


Figure S1. XRD spectrum of the as-synthesized CdSe nanobelts.

S2. Structural comparison between CdSe source powder and nanobelts by XRD

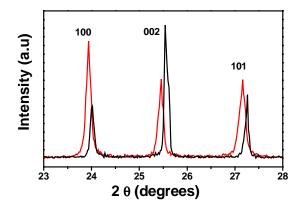


Figure S2. XRD spectra of CdSe source powder (red line) and the

as-synthesized CdSe nanobelts (black line).

S3. An intermediate state of CdSe at ~1.43 eV

In our SERS measurements of single CdSe nanobelts excited at 785 nm with laser polarization parallel to the c-axis ($||_c$) of CdSe nanobelt, a weak broad emission band at ~1.43 eV was observed (Figure S3a). In sharp contrast, this emission band disappeared when the polarization of the excitation light was perpendicular

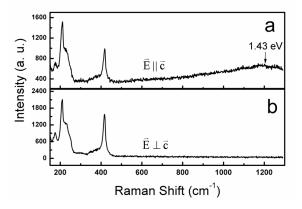


Figure S3. SERS spectra of a single CdSe nanobelt excited at 785 nm with laser polarizations (a) parallel and (b) perpendicular to the c-axis of CdSe nanobelt.

to the c-axis (\perp_c) of CdSe nanobelt (Figure S3b). The dipole polarization (\parallel_c) of the intermediate state at ~1.43 eV, lower in energy to the A exciton (~1.74 eV) of CdSe, is different from that (\perp_c) of the A exciton.

Broad emission bands at ~1.4–1.7 eV were observed in CdSe thin films in several previous studies.^{S1-S3} These bands have been assigned as intermediate states (donor acceptor levels) due to defects (Se vacancies) or impurity contaminations (Cl, Na, and K) in CdSe crystals.^{S2,S3} In our experiments, no contaminations or impurities could be detected by XEDS (X-ray energy-dispersive spectrometer). The weak emission band at

~1.43 eV observed in Figure S3a might suggest a slight deviation of stoichiometric ratio of 1 : 1 between Cd and Se in the as-synthesized CdSe nanobelts.

S4. Laser power dependence of the SERS intensities of CdSe nanobelts

Laser power dependence of the SERS intensity for single CdSe nanobelts excited at 785 nm is conveyed in Figure S4a. Increments of the SERS intensities for different phonon modes with increasing laser power are shown in Figure S4b. All of these phonon modes show linear power dependences in the laser intensity range of ≤ 70 kW/cm². In the increment of laser power, an assumption of the rise of local temperature at nanobelts is reasonable. Figure S4c shows the intensity ratio $\zeta = I_{2LO}/I_{LO}$ of a CdSe nanobelt increases slightly from 0.43 to 0.57 with increasing laser power. This

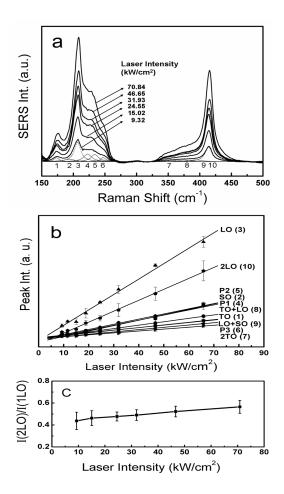


Figure S4. (a) SERS spectra of a single CdSe nanobelt excited at 785 nm with different laser intensities. Phonon modes (marked 1–10) deconvoluted from the bottom spectrum are labeled parallel to those in Figure 3. (b) Power dependence of the deconvoluted phonon modes. (c) Power dependence of $\zeta = I_{2LO}/I_{LO}$.

observation agrees with the theoretical prediction in a previous study of CdSe nanospheres,^{S4} where theoretical ζ increases with increasing temperature, provided the dephasing rate ($\Gamma = 1/T_2$) in the CdSe crystalline system is thermally insensitive to the Raman excitation.^{S5}

References

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(S5) Raman cross section for the nth order of the LO mode in CdSe can be calculated using Eq. (49) of Ref. S4.