Supporting Information

Ion transport induced room-temperature insulator-metal transition in single-crystalline Cu_2Se

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Crystal growth conditions

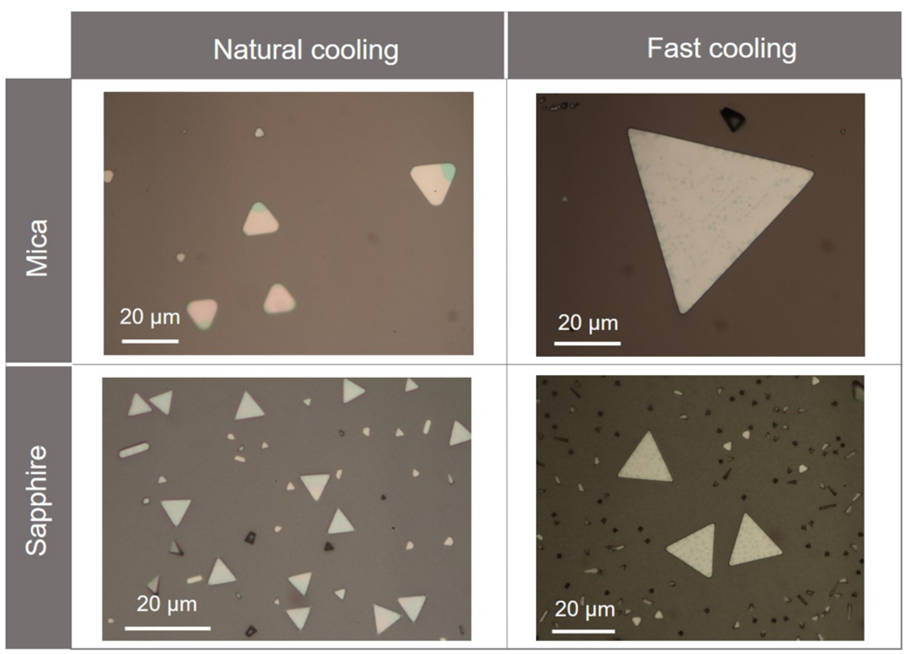


Figure S1. Optical micrographs of Cu_2Se crystals grown on mica and sapphire substrates under different cooling rates.

SEM and AFM images

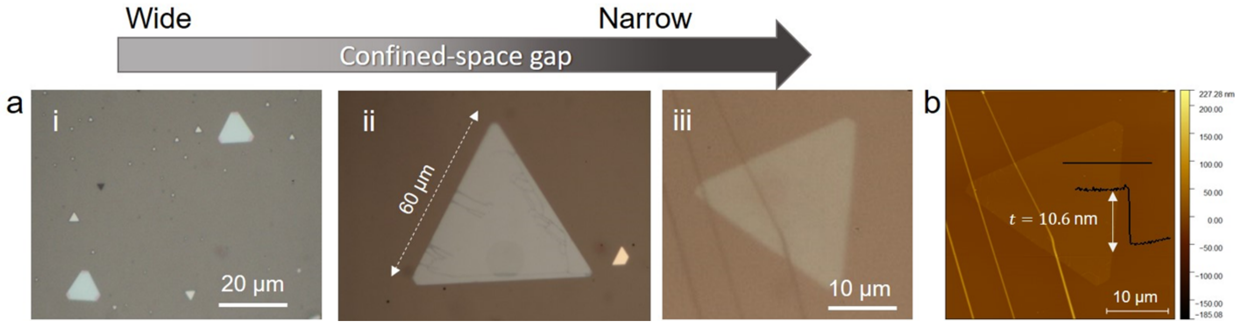


Figure S2 a. Optical micrographs of Cu_2Se crystals synthesized on mica substrate via a confined-space approach. **b.** AFM height trace map of an 11 nm thick Cu_2Se crystal.

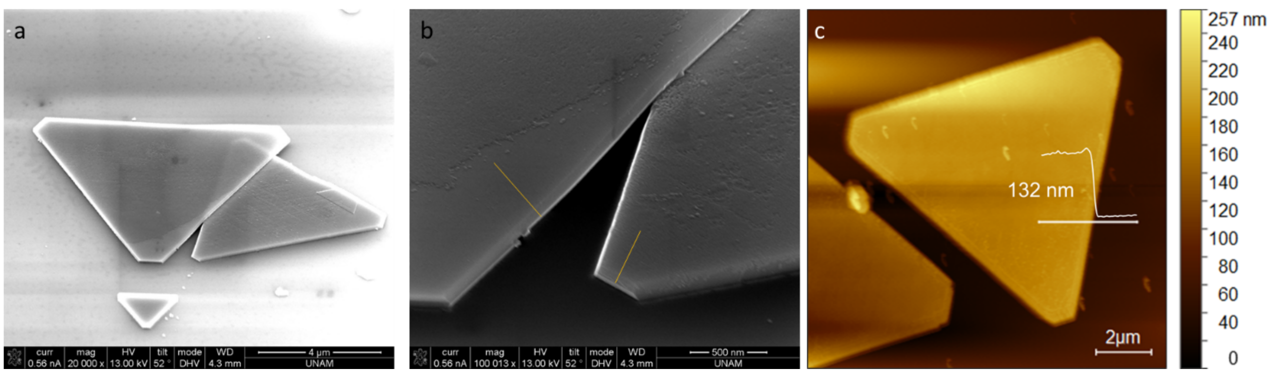
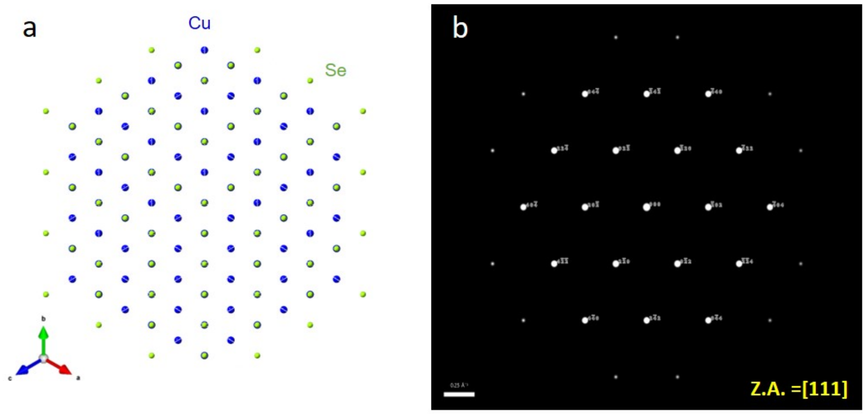


Figure S3 a. SEM micrograph of crystals transferred over SiO_2/Si substrate and **b.** close-up view of the crystal edges. Yellow dashed lines indicate the wedge-shaped edges of the crystal. **c.** AFM image of a Cu_2Se crystal with a thickness of 150 nm grown on a mica substrate. Crystal's wedge-shaped edges are visible in the AFM maps.

Simulated SAED Pattern



Chemical Formula: Cu_2Se
Space Group: $Fm\bar{3}m$
Crystal System: Cubic
a = 4.103 Å
b = 4.103 Å
c = 4.103 Å

h	k	l	d [nm]
-2	2	0	0.205132
-2	0	2	0.205132
0	-2	2	0.205132

Figure S4. Atomic arrangement and simulated SAED pattern of cubic Cu_2Se.

Temperature cycling of Cu_2Se under vacuum

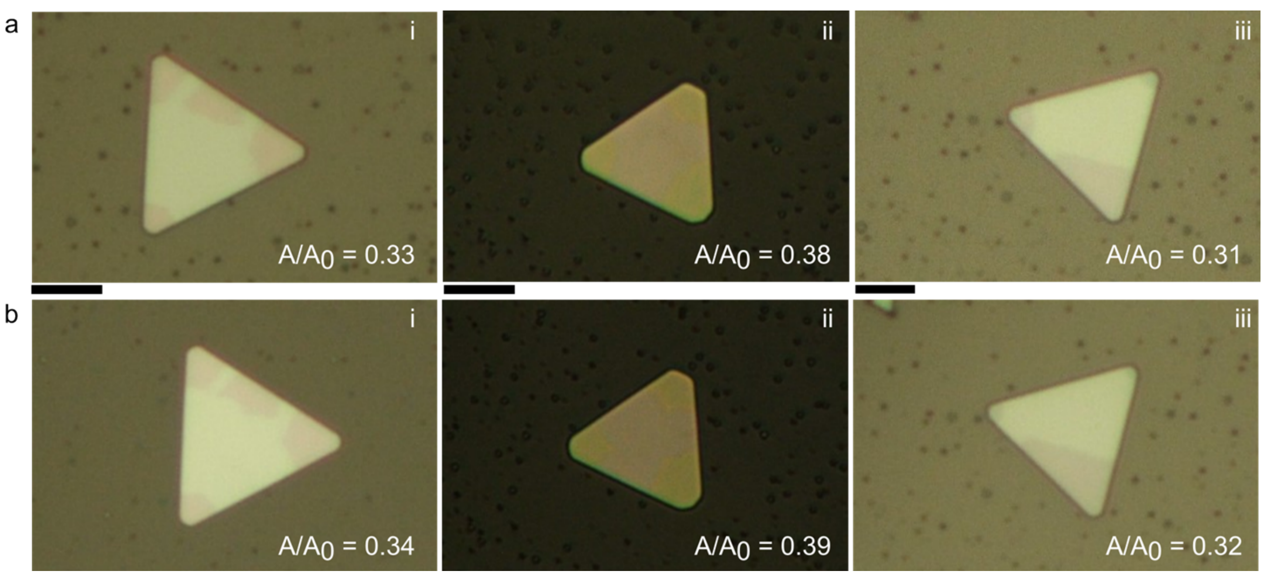


Figure S5 a. Optical micrographs of various Cu_2Se crystals taken before temperature cycling to 400 K are given. **b.** Optical micrographs of the same crystals at room temperature after heating to 400 K under 10^{-2} mbar vacuum. A/A_0 ratio is indicated in the figures. The scale bars for panels i and ii are 10 μm and for iii 5 μm.

Temperature cycling under ambient and laser manipulation of the dark phase

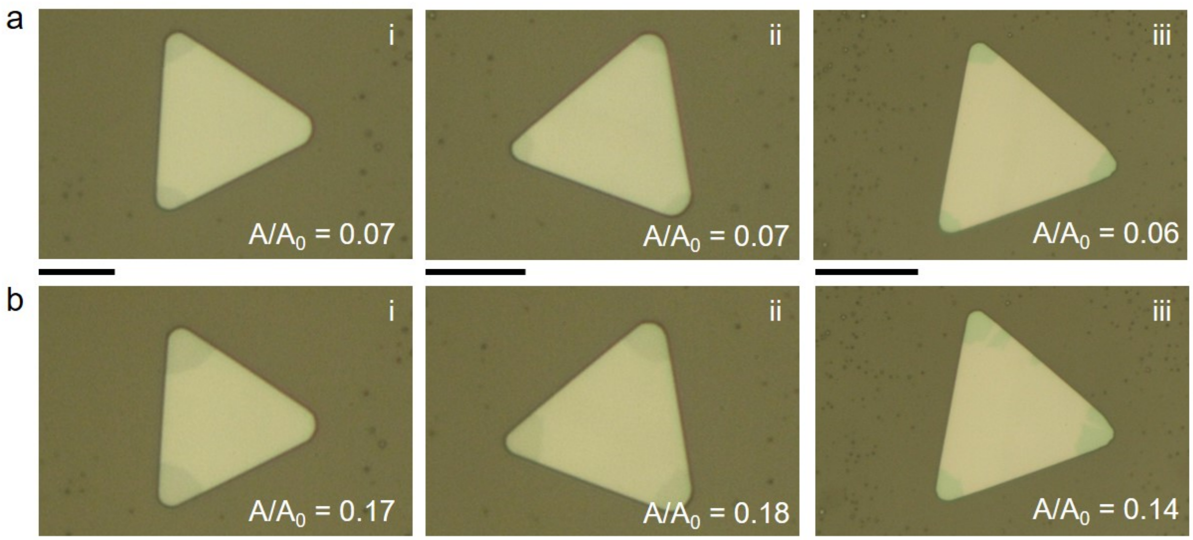


Figure S6 a. Optical micrographs of various Cu_2Se crystals taken before temperature cycling from room temperature to 400 K. **b.** Optical micrographs of the same crystals at room temperature, after heating to 400 K under ambient. A/A_0 ratio is indicated in the figures. The scale bars for panels i and ii are 10 μm, and for iii is 20 μm.

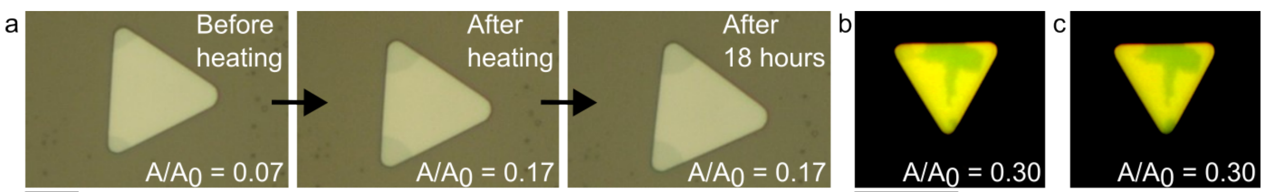


Figure S7 a. A series of optical micrographs of Cu_2Se crystal before heat cycling under ambient, after heat cycling above 400 K, and after keeping it at room temperature in ambient for 18 hours is shown. As discussed in the main text, A/A_0 ratio increases after heat cycling and remains unchanged at room temperature. **b.** Optical microscope micrograph of a crystal at 348 K before laser manipulation and **c**. after laser manipulation. A/A_0 ratio remains the same. The scale bars are 10 μm.

Laser illumination-induced phase transition

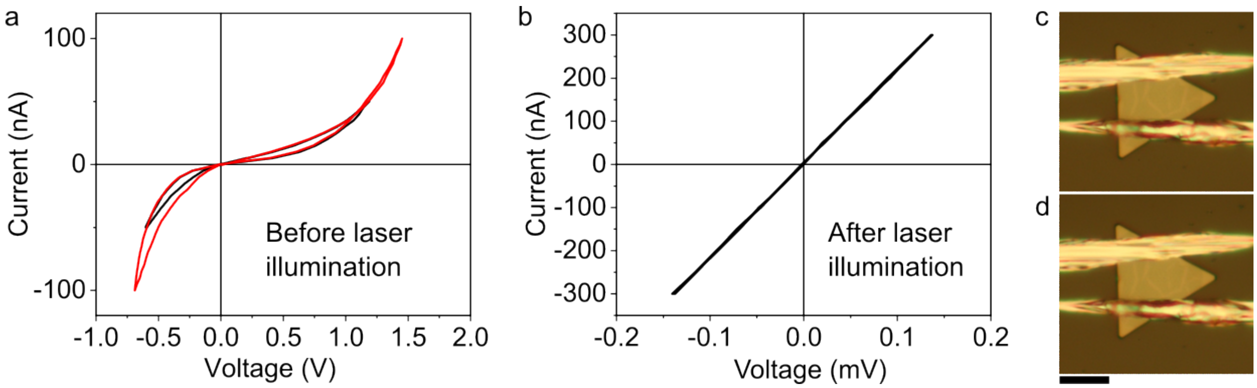


Figure S8 a. I-V curves of an indium-contacted Cu_2Se device before laser illumination and **b.** I-V graph of the same device after laser illumination. 212 μW laser power is used to illuminate the crystal partially. **c.** Optical micrograph of the device before illumination and **d.** after illumination is given. The scale bar is 10 μm.

Copper leakage to electrodes

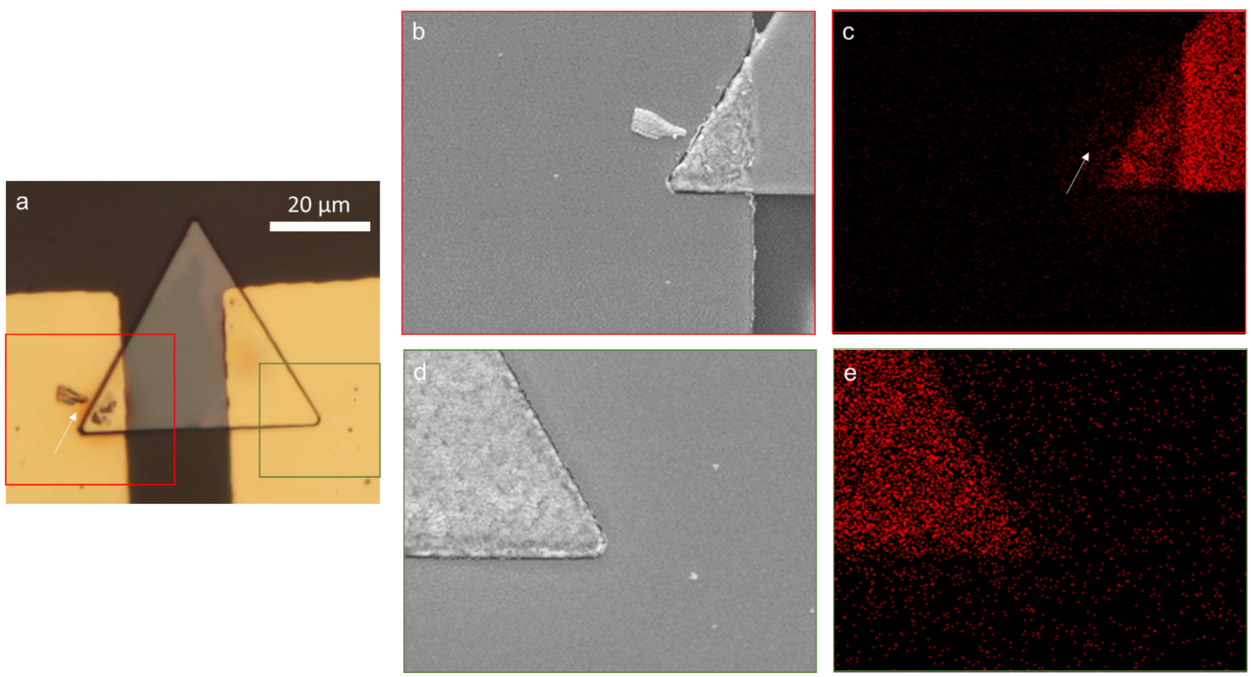


Figure S9 a. Optical microscope image of a two-terminal device after becoming conductive. The arrow points to the red color on the gold electrode, formed after Cu depletion. **b.** SEM image of the left contact, marked by the red rectangle in **a** and **c.** corresponding EDAX map of Cu-L. The white arrow shows the copper accumulated regions in the contacts. **d.** SEM image of the right contact, marked by the green rectangle in **a** and **e.** corresponding EDAX map of Cu-L. Only the signal is from the corner of the crystal, and the uniformly distributed signal from the rest of the image is due to background noise.

Temperature-dependent resistance at higher temperatures

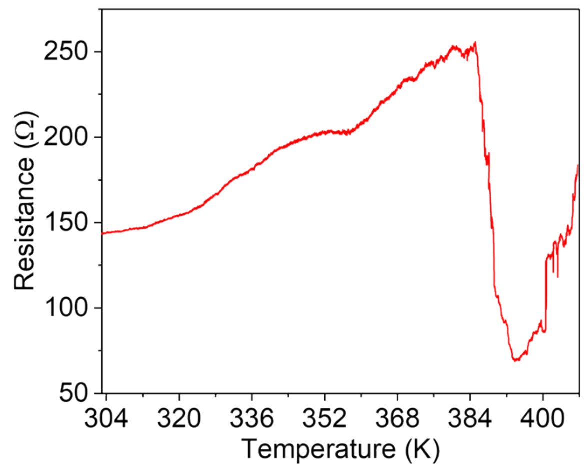


Figure S10. The temperature-dependent electrical resistance of a two-terminal Cu_2Se single crystal at higher temperatures.