Effects of resonant bonding and structural distortion on phase change properties of Sn$_2$Sb$_2$Se$_5$

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S1. Homogeneity of crystalline SSS film.

Figure S1. HRTEM image and EDS results of crystalline SSS film. The red, green, and blue dots represent individual atoms.
S2. Bond length distributions of Sn-Se bond and Sb-Se bond in crystalline SSS

Figure S2. Calculation results showing radial distributions of Sn atoms and Sb atoms around Se atoms in crystalline SSS.
S3. Phonon dispersion and Raman mode analysis results for crystalline SSS

Figure S3. Calculation results for phonon dispersion of crystalline SSS in case of six-fold Sn atom.
Figure S4. Schematic diagram of Raman vibrational mode in SSS simulated from calculation result in Figure S3. C, D, and E correspond to each peak of the Raman spectrum in Figure 2(b).

Generally, the peaks in Raman spectra originate from the vibration modes of the structure, which differ based on the bonding characteristics and structural differences. In order to predict Raman vibration modes C, D, and E in Figure 2(b) to obtain additional atomic structural information, phonon calculations with DFT were performed using the calculated crystal structure of SSS. The phonon dispersion of SSS was calculated from its structure in each case of 0-fold to 6-fold Sn atoms. The specific vibrational modes were determined by simulating and analyzing the vibrational direction of the atoms in each case of Sn atoms at the local minimum point in the phonon dispersion. The specific vibrational modes were matched to the vibrational modes obtained in Figure 2(b) by calculating the corresponding Raman frequency using the phonon dispersion results. As a result, vibrational mode C, D, and E in the figure were determined to be the $E_{2g}$ bending, $A_{1u}$ stretching, and $A_{1u}$ stretching modes, respectively.