

SUPPORTING INFORMATION

Parallel Exploration of the Optoelectronic Properties of (Sb,Bi)(S,Se)(Br,I) Chalcohalides

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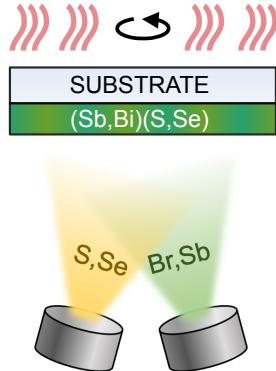
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- 1. Precursors:** Co-evaporation from elemental pnictogen (Sb, Bi) and chalcogenide (S, Se) sources



- 2. Chalcohalides:** High-pressure reactive annealing of binary precursors with pnictogen halides (BiI_3 , BiBr_3 , SbI_3 , or SbBr_3)

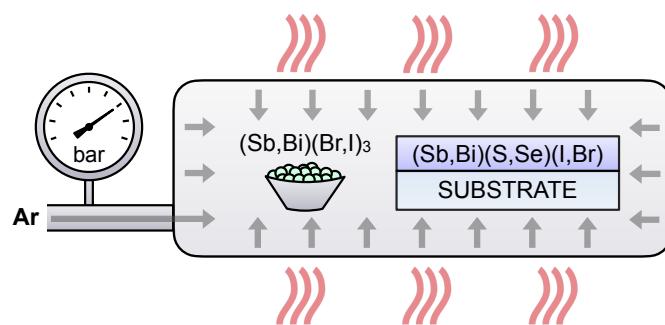


FIG. S1. Schematic illustration of the two-step physical vapor deposition (PVD) process used to synthesize pnictogen chalcohalides. In the first step, a binary precursor thin film is deposited via thermal co-evaporation of elemental pnictogen and chalcogenide sources. In the second step, the precursor film is placed in a pressurized furnace with pnictogen halide source materials, where high-pressure reactive annealing converts the film into the desired ternary chalcohalide phase.

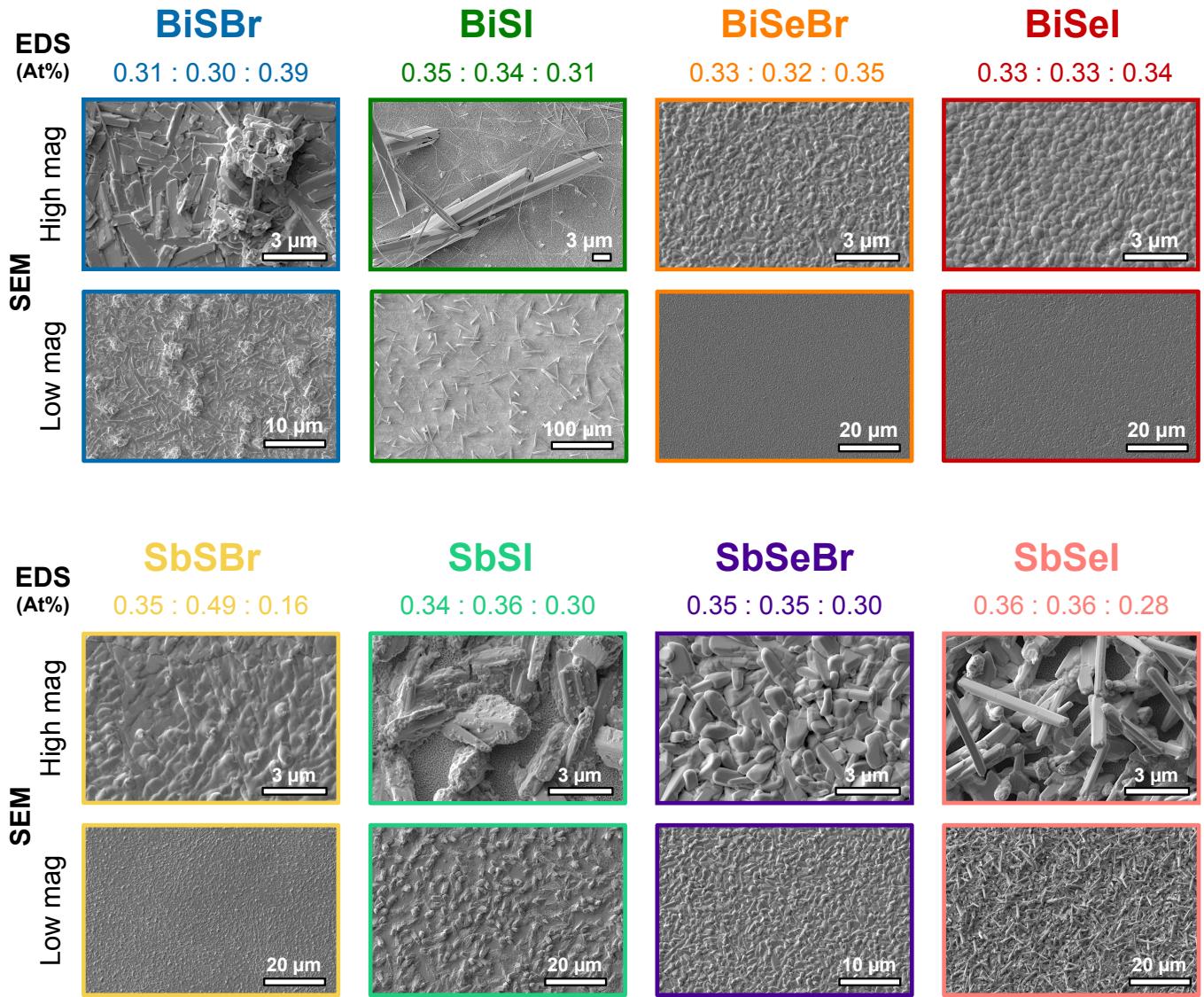


FIG. S2. Top view SEM images at high and low magnification for all Bi- and Sb-based chalcohalides, along with corresponding atomic percentages from EDS analysis.

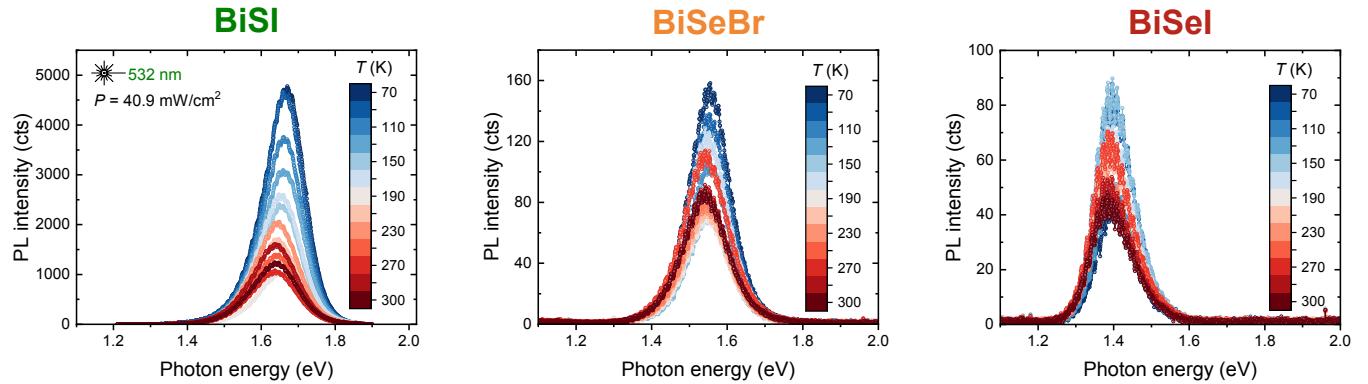


FIG. S3. Temperature-dependent PL spectra of BiSI, BiSeI, and BiSeBr from 70 to 300 K under 532 nm excitation at 40.9 mW/cm².

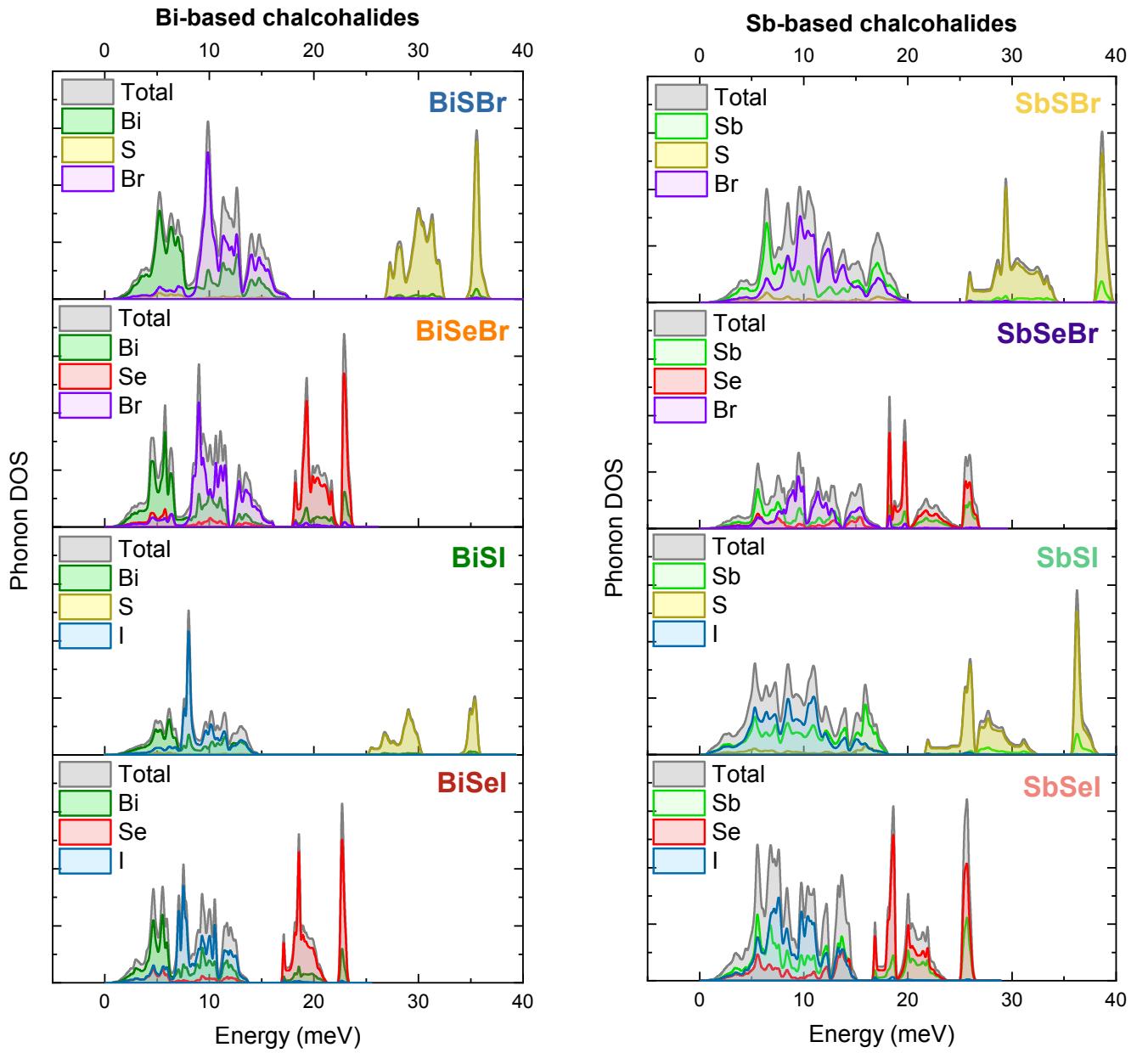


FIG. S4. Projected phonon density of states (DOS) for all Bi- and Sb-based chalcohalides. All sulfur-based compounds exhibit a phonon gap below the high-frequency chalcogen band, while selenium-based compounds show Se-related modes that fill this gap, enhancing phonon scattering pathways.

TABLE I. Room-temperature bandgap energies for all eight chalcohalide compounds, determined from the photoluminescence peak positions. The value for SbSeBr is marked with an asterisk (*) to indicate that the structural determination for this compound was inconclusive; therefore, the observed deviation from the sulfur-selenium trend should be interpreted with caution.

Compound	E_g (eV)
BiSeI	1.38
BiSeBr	1.54
BiSI	1.64
BiSBr	1.99
SbSeI	1.74
SbSBr	1.81
SbSI	1.91
SbSeBr*	2.08