Supporting Information

Photoresponse of Natural van der Waals Heterostructures

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Energy Dispersive Spectroscopy (EDXS) on the SEM of Franckeite

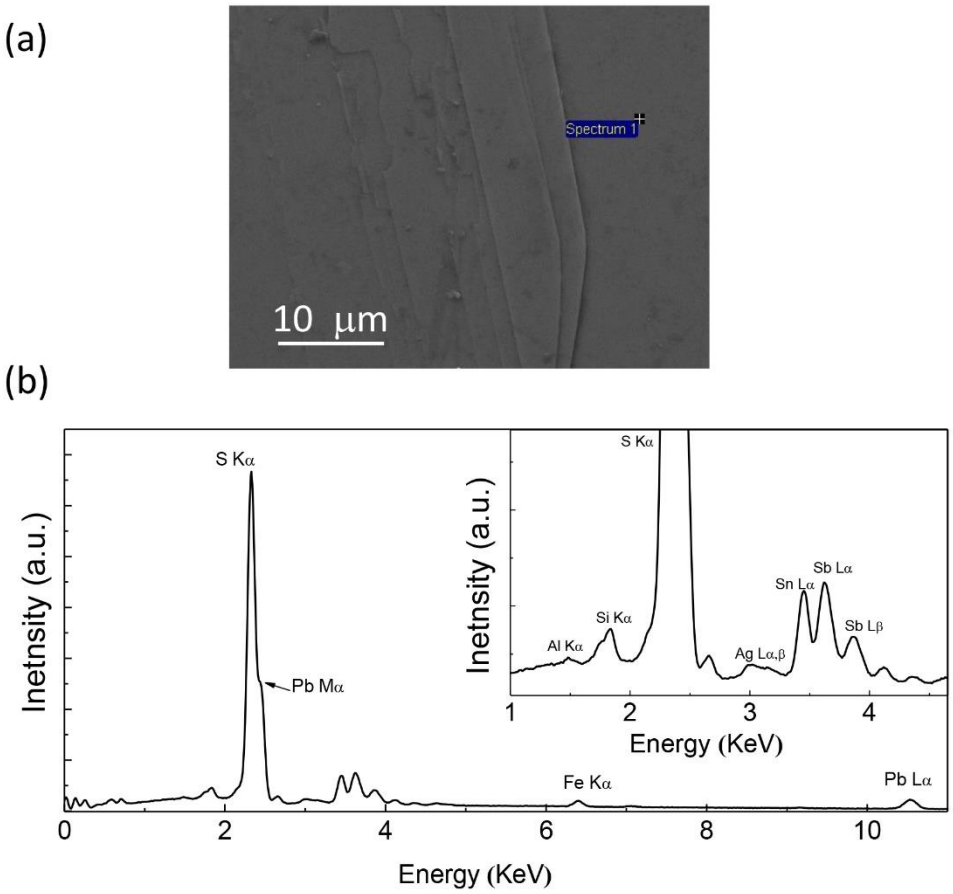

Figure S1: SEM and EDX characterization of franckeite. (a) SEM image of a franckeite flake used for the EDX study. (b) Average EDX spectra obtained for the sample.

Element	Concentration (at %)
Al	0.61
S	57.49
Fe	3.43
Sn	9.59
Sb	8.88
Pb	19.02
Ag	0.98
Totals	100

Table 1: EDXS quantification of franckeite crystal.

Photoresponse of franckeite samples.

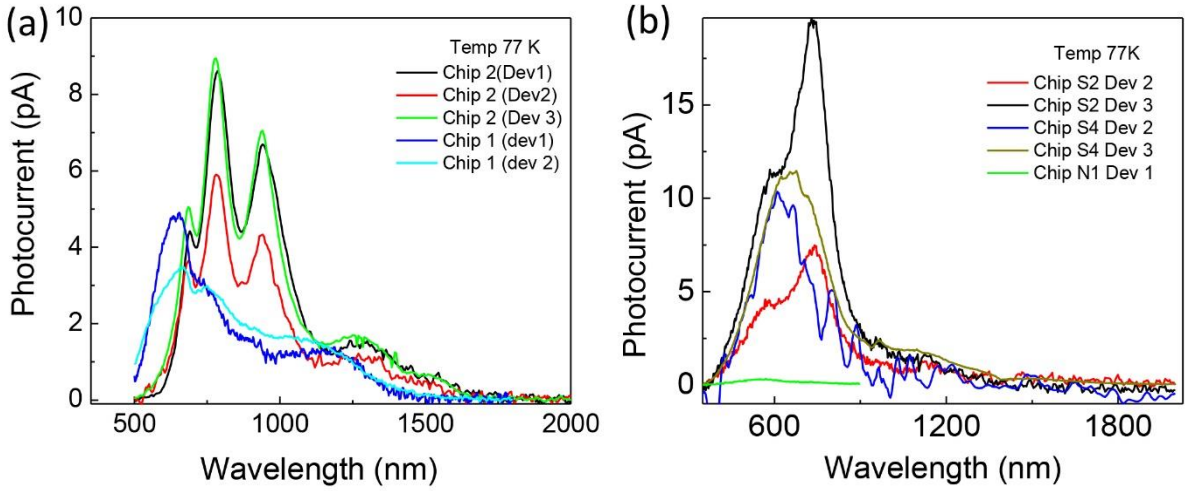

Figure S2: The PC response of 10 devices based on franckeite crystals. The measurement was conducted at 77K. Note that some peaks are due to the source spectrum.

DFT Calculations

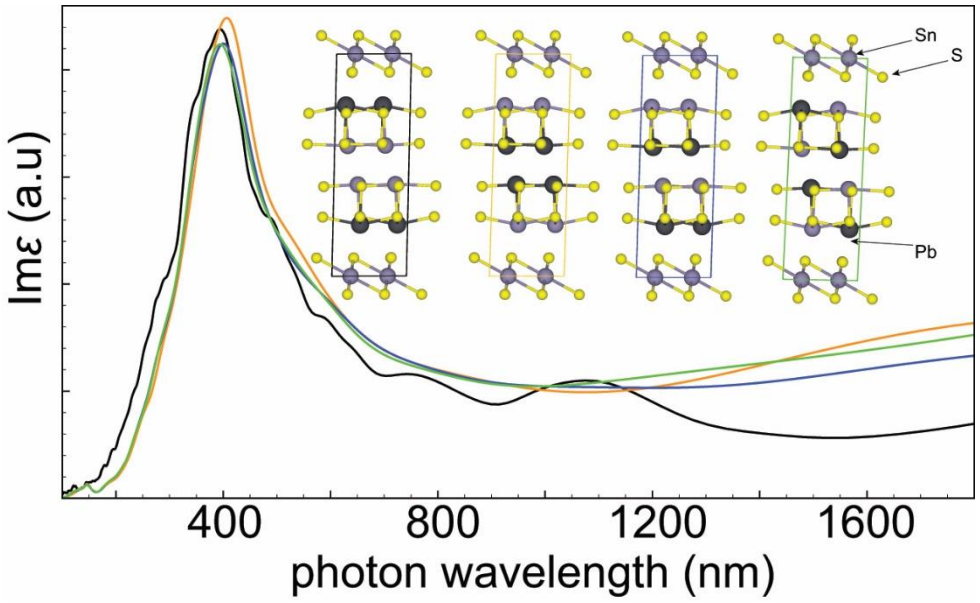

Figure S3: DFT-PBE calculated imaginary part of the dielectric function of the Franckeite bulk structure with varied stacking (insets) in the Q layer.

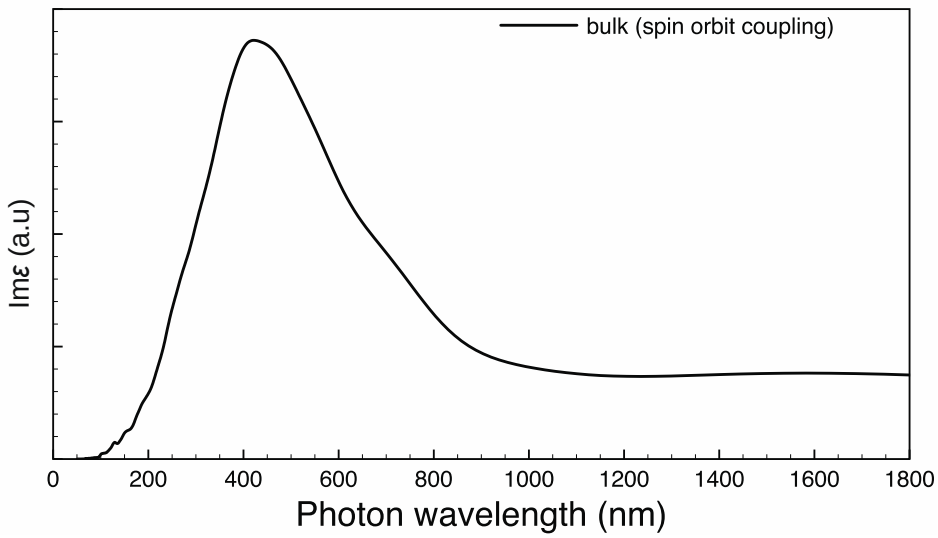

Figure S4. The imaginary part of the dielectric function for the franckeite bulk material with spin orbit coupling included.

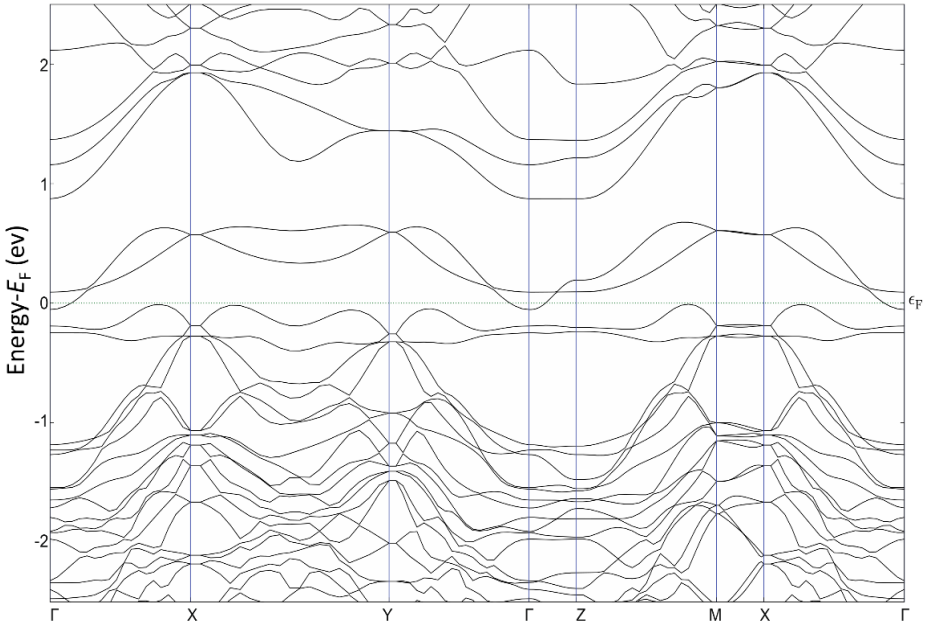

Figure S5. DFT calculated band structure of the bulk franckeite with spin-orbit coupling included

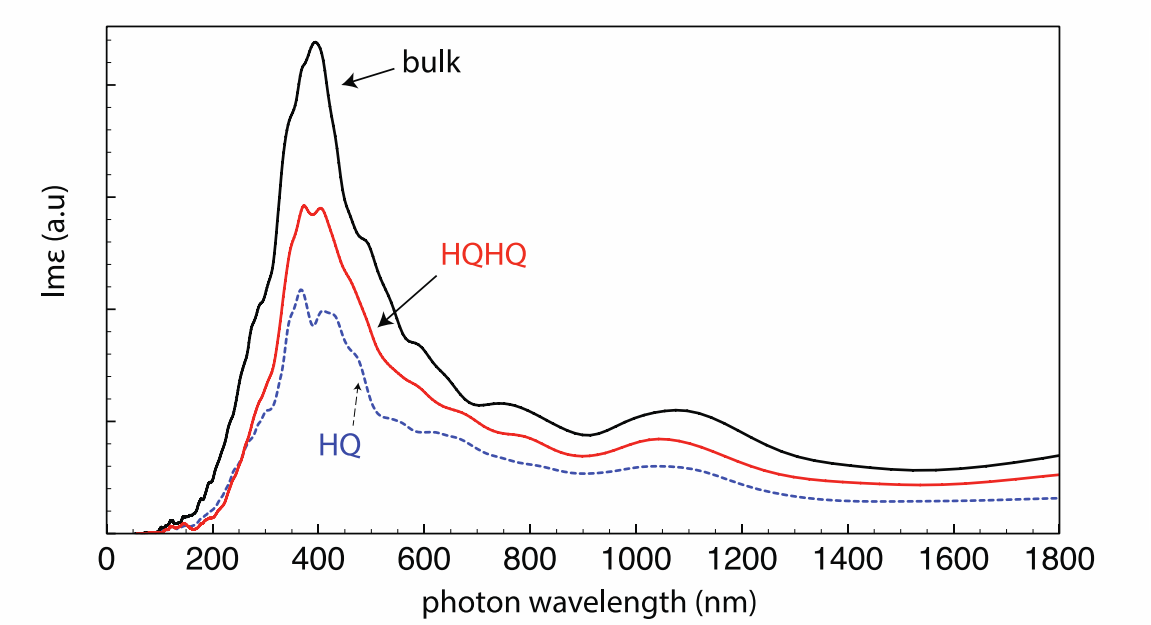

Figure S6. Calculation of the optical absorption properties of franckeite. The imaginary part of the dielectric function of HQHQ layer (red solid) and HQ layer (blue dashed) as compared with the bulk franckeite (solid black).

Thickness measurement

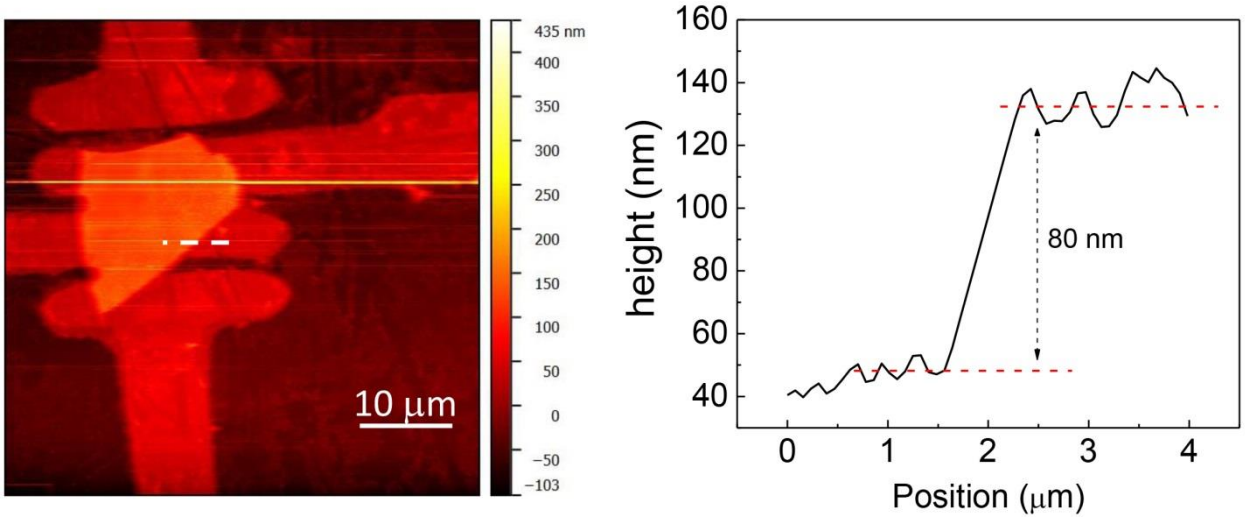

Figure S7: Thickness measurement by AFM. (Left) An AFM image of the flake on Au electrodes taken after the dry transfer. (b) The line profile of the sample along the white dashed line in the left figure.