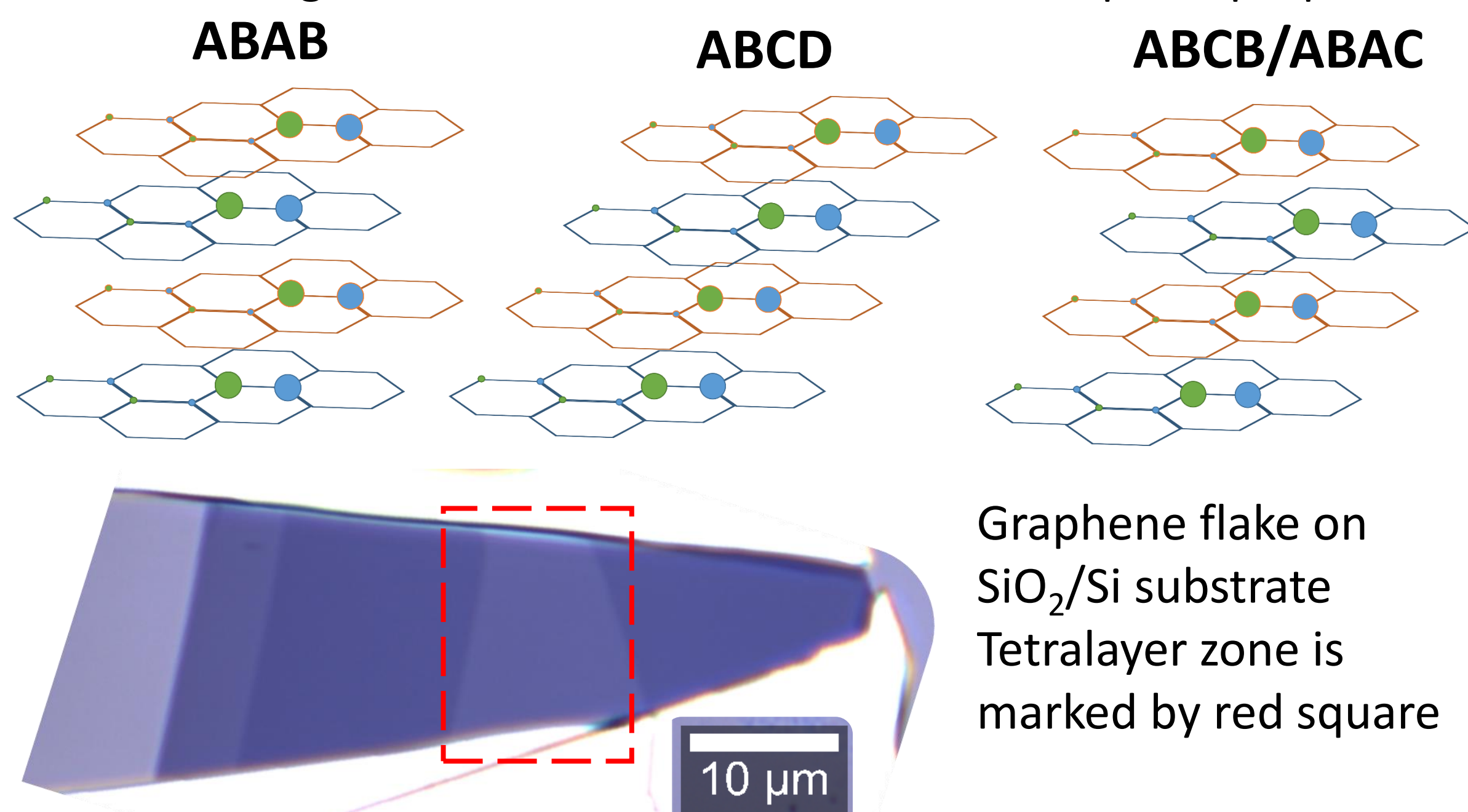


Abstract

The stacking order of Few-Layer Graphene (FLG) affects its electronic band structure, altering its optical properties. Tetralayer graphene has three possible stacking phases: two were experimentally observed Bernal (ABAB) and Rhombohedral (ABCD), while the elusive third ABCB/ABAC has proven much harder to detect. In this work, we show that single wavelength (0.63 eV) Scattering Scanning Near-field Optical Microscopy (S-SNOM), in combination with an Oscillating Spherical Dipole (OSD) model, can be used to clearly image and categorize all three phases of Tetralayer graphene. Furthermore, we show that the method may be used for imaging larger-order stacking graphene.

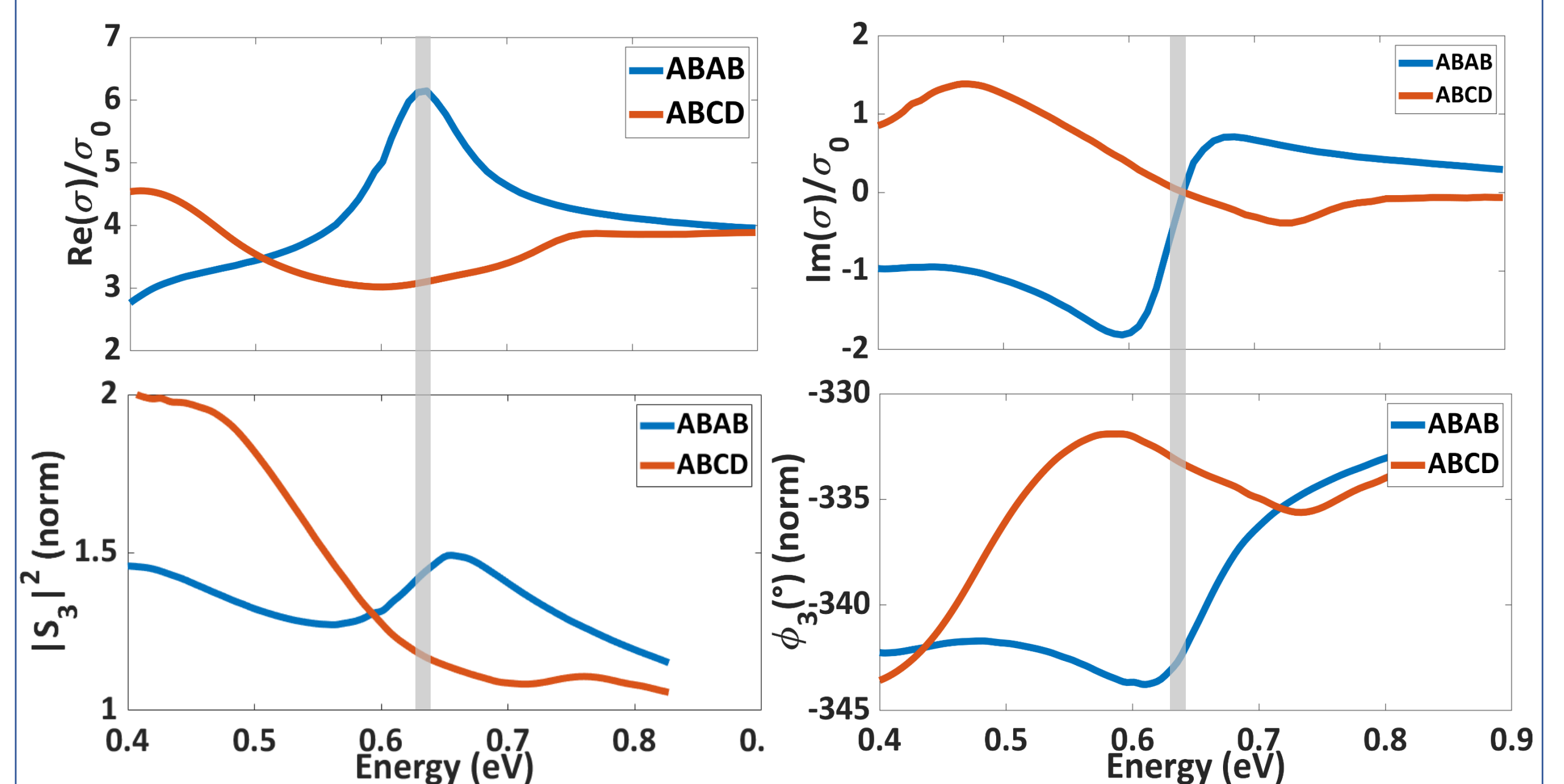
Tetralayer Graphene stacking order

- In tetralayer graphene there are 3 different stacking options
- The ABCB/ABAC stacking are identical except inverted
- Each stacking has a different band structure and optical properties



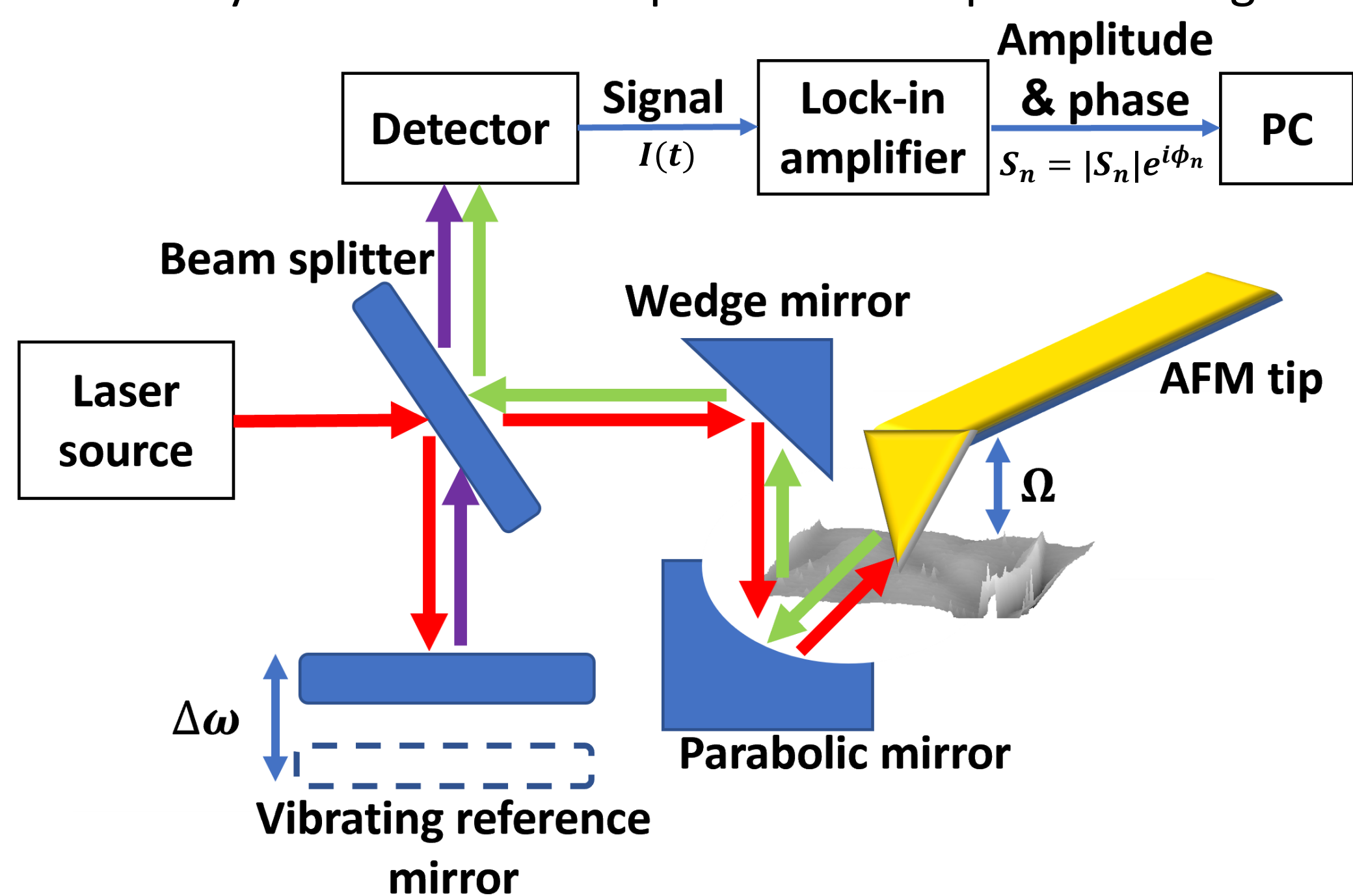
Tetralayer Graphene S-SNOM contrast

- Theoretical spectra of optical conductivity (normalized by $\sigma_0 = c\alpha/4$)¹
- Calculated S-SNOM third harmonic signal - $S_3 = |S_3|e^{i\phi_3}$
- At 0.63 eV (marked by shaded area) $|S_3(ABAB)| > |S_3(ABCD)|$ and $\phi_3(ABCD) > \phi_3(ABAB)$

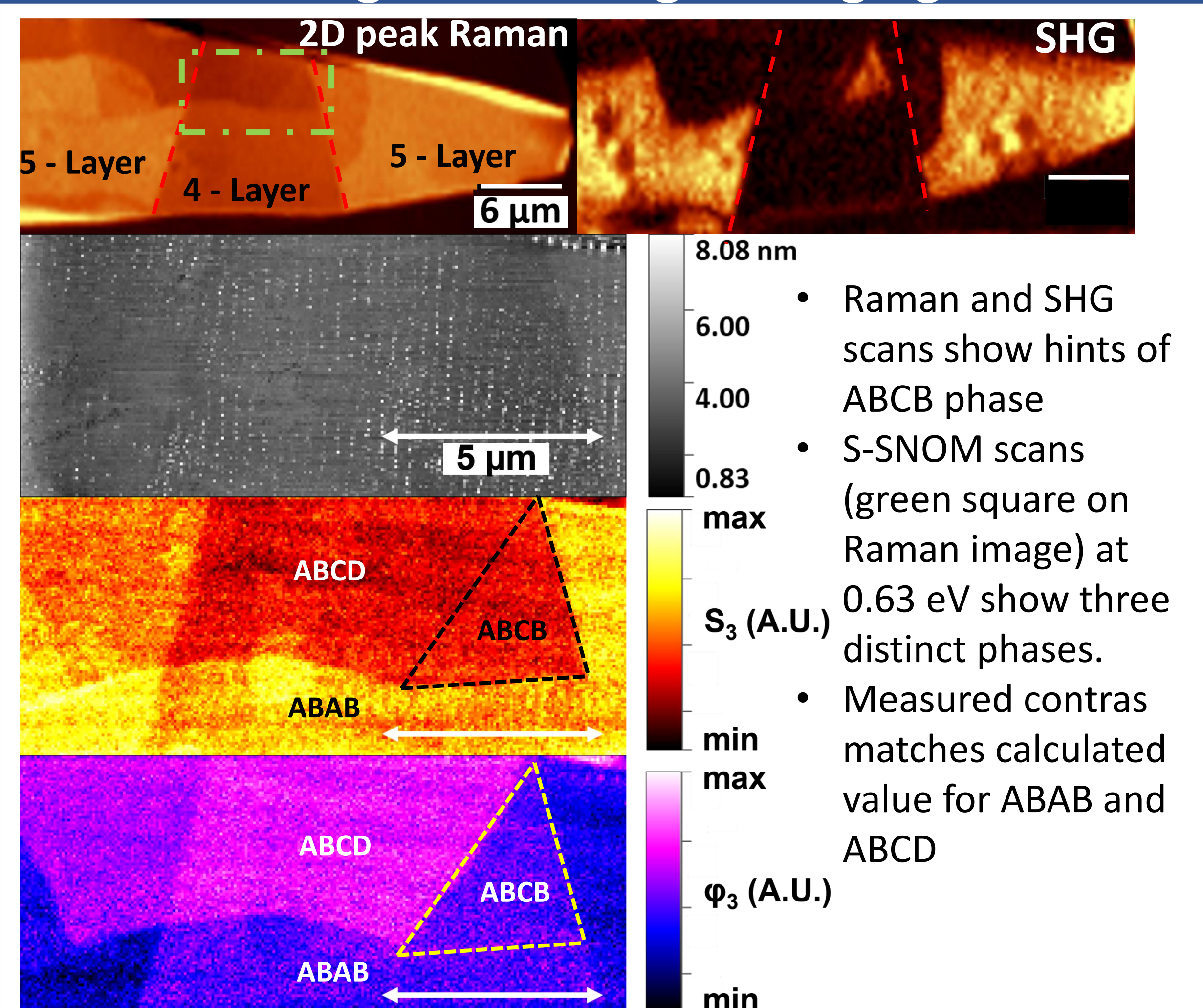


S-SNOM

- Metallic tip focuses light beyond the diffraction limit (res < 30 nm)
- Lock-in Amplifier extracts near-field signal in higher harmonics
- PS- Heterodyne retrieves both phase and amplitude of signal

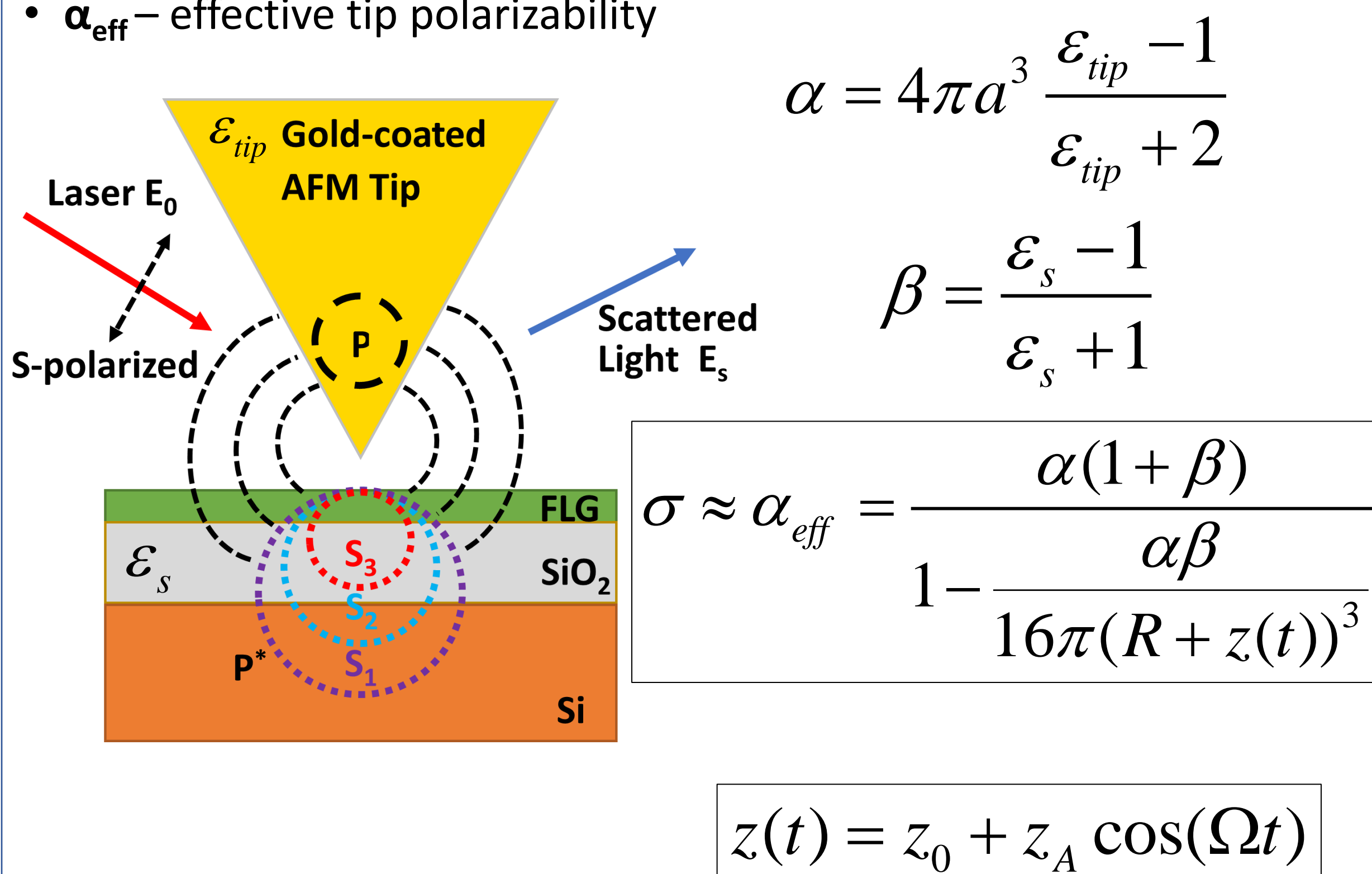


Single Wavelength Imaging



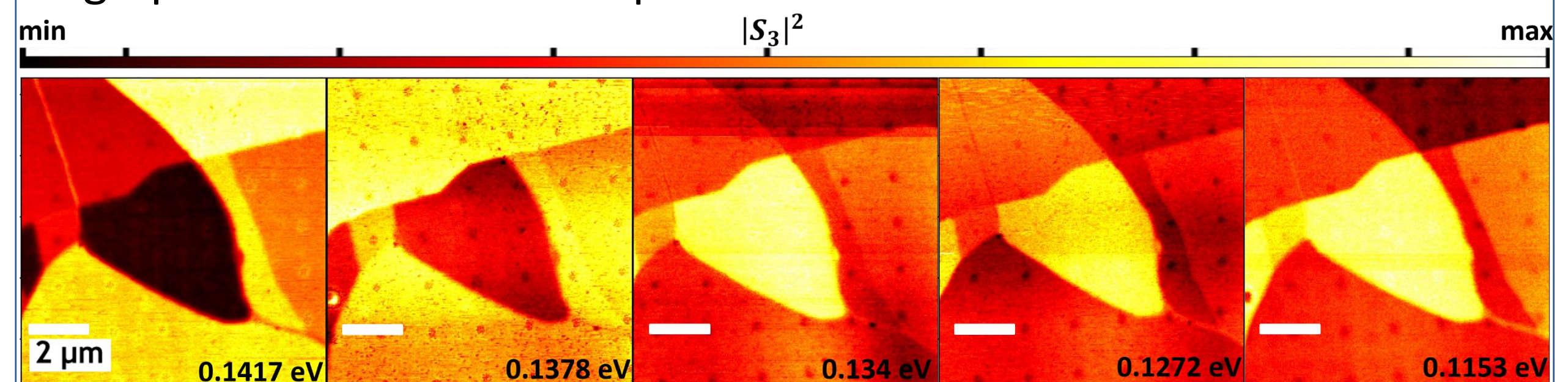
Oscillating Spherical Dipole Model

- The incoming laser induces a dipole \mathbf{P} on the metallic tip
- The metallic tip induces an image dipole \mathbf{P}^* in the material
- interaction between the dipoles is by the reflection parameter β
- α – polarizability for a Spherical metallic dipole
- α_{eff} – effective tip polarizability



Higher layer number stackings

- Variable contrast in Mid-IR measurements of higher layer number graphene reveal different phases



Conclusions

- All three possible stacking phases of Tetralayer Graphene were imaged via S-SNOM
- Single wavelength imaging is sufficient to detect and categorized different phases in FLG
- Higher layer number graphene reveal variable contrast behavior in Mid-IR