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Correlating nonlinear optical properties with atomic-level structure in 2D-polar metals

We report on the nonlinear optical properties of 2D polar metals, an emerging group of materials synthesized through confinement heteroepitaxy (CHet) [1]. These heterostructures, consisting of two- to three- atomic layers of indium or gallium on a SiC substrate with a graphene capping layer, exhibit promising nonlinear optical properties in the technologically important NIR/Vis frequency range [2]. The large second order nonlinear susceptibility, approaching 10 nm/V, is unexpected for a centrosymmetric material. However, sub-Ångstrom changes in the lattice parameters as the bonding evolves over the thickness of the metal break the centrosymmetry and result in an out-of-plane dipole, which is confirmed by angle-resolved SHG microscopy. While the out-of-plane structure allows the second-order nonlinear response, the atomic-level in-plane structure of the 2D-metal imparts unique polarization-dependent nonlinear responses. Using polarization-resolved SHG microscopy, we identify regions which have small differences (<2Å) in the lateral displacement of successive layers of metal due to step edges in the SiC substrate. Both the in-plane and out-of-plane structure is determined by the strong interaction of the metal with the SiC substrate, suggesting the ability to tune polarization-selective nonlinear optical properties by control of the interface. Understanding of the correlation between structure and nonlinear optical responses of 2D-metals in an air-stable platform may enable future breakthroughs in nonlinear optical technology.

References

[1] N. Briggs et. al., Nat. Mat., 19 (2020) 637-643.
[2] M. A. Steves et. al. Nano Let., 11 (2020) 8312-8318.

Figures

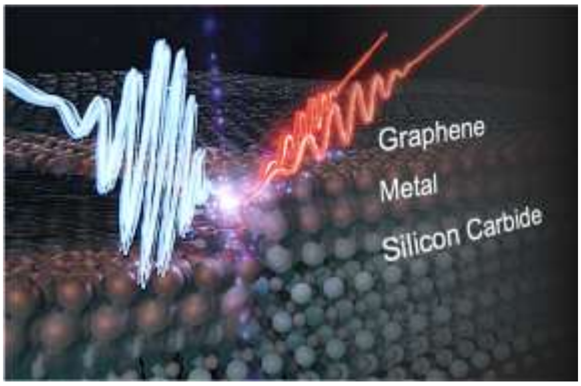

Figure 1: Second harmonic generation microscopy of 2D-polar metal heterostructure