Nonlinear Optical Behavior in Two-Dimensional Polar Metals

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Abstract: Nonlinear optical materials, materials with a nonlinear dependence on the incident field strength, are utilized in telecommunications and pulsed laser technologies due to their ability to modify the frequency, phase, and amplitude of light. Prior work has demonstrated that two-dimensional polar metal heterostructures (PMets), which feature a few-atomic-layer crystalline metal film intercalated in the confines between silicon carbide and a graphene capping layer, efficiently transduce second harmonic generation (SHG). The efficient SHG is due to the highly polarizable metal-SiC interface; however, other nonlinear optical behavior such as enhanced SHG due to resonance matching and high-order harmonic generation (HHG) have not been explored. Herein, we demonstrate that the behavior of SHG from 2D Ag is demonstrated to be structure-dependent while the HHG of graphene can be modified by the substrate-metal interface. 2D Ag demonstrates domain-specific behavior, which is currently assigned to two different structural motifs of the intercalated Ag. The power dependence of steady-state and interferometric SHG verify that both saturable and enhanced SHG behaviors are present. The saturable SHG is similar to prior simulations of interferometric SHG which indicates that a population inversion can be induced by resonant excitation of an excited state. In a separate project, 2-D PMets demonstrate the ability to produce HHG, which is believed to be sourced by the graphene. In most systems, HHG emission rapidly reduces as the fundamental light becomes increasingly circular; however, graphene has a unique elliptical signature which we use to identify the source of HHG. We hypothesize that the HHG signal from PMets is dominated by the graphene which is modified via the work function of the graphene-substrate interface.