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Article

Efficient High-Order Harmonic Generation from the van der Waals Layered Crystal Copper Indium Thiophosphate

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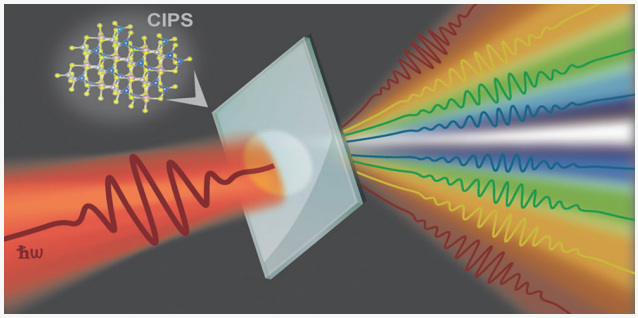
ABSTRACT: Layered metal thio- and selenophosphates (MTPs) are a family of van der Waals gapped materials that exhibit a multitude of functionalities in terms of magnetic, ferroelectric, and optical properties. Despite the recent progress in terms of understanding the material properties of these compounds, the potential of MTPs as a material class yet needs further scrutiny, especially in terms of nonlinear optical properties. Recent reports of efficient low-order harmonic generation and extremely high third-order nonlinear optical properties in MTPs suggest the potential application of these materials in integrated nanophotonics. In this article, we investigate the high-order nonlinear response of bulk and exfoliated thin-film crystals of copper indium thiophosphate (CIPS) to intense mid-infrared fields through experimental and computational studies of high-order harmonic generation (HHG). From a driving laser source with a 3.2 μm wavelength, we generate odd and even harmonics up to the 10th order, exceeding the bandgap of the material. We note conversion efficiencies as high as 10^{-7} measured for the fifth and seventh harmonics and observe that the harmonic intensities follow a power law scaling with the driving laser intensity, suggesting a perturbative nonlinear optical origin of the observed harmonics for both bulk and thin flakes. Furthermore, first-principles calculations suggest that the generation of the highest harmonic orders results from electron−electron interactions, suggesting a correlation-mediated enhancement of the high-order optical nonlinearity.



INTRODUCTION

The strong-field nonlinear response of a material under the influence of an intense, low-frequency femtosecond laser field is progressively being exploited for the high-order harmonic generation (HHG) of coherent high-frequency light. HHG harnesses the electron dynamics driven by intense, low-frequency driving lasers to produce high-energy photons which can be applied for time-resolved spectroscopy, metrology, or imaging applications.[1] As the emission of HHG from solids is a consequence of microscopic laser-driven currents, the same physics can be harnessed for the development of lightwave electronics devices operating at optical frequencies.[2,3]

HHG and other strong-field processes have been well studied in atomic and molecular gases.[4] More recently, long-wavelength driving laser fields in the mid-infrared spectral region have enabled the experimental observation of HHG from solid-state samples.[1,5,6] The advancement of high-order harmonic spectroscopy and the application of intense, low-frequency fields in novel optoelectronic devices necessitate the exploration of new nonlinear optical (NLO) materials with optical and chemical stability to generate coherent short-wavelength light sources and to convert long-wavelength laser light into petahertz-frequency electronic signals. Conventional nonlinear photonic devices which are based on bulk lithium niobate ($LiNbO_3$) and barium titanate ($BaTiO_3$) pose limitations such as low nonlinear susceptibility and phase matching issues which make them ill-suited for future nonlinear photonic devices.[6] While HHG in solids stems from a weaker polarization response than low-order nonlinearities, it is a near-universal phenomenon that can be realized in a wide array of material systems without phase matching, and therefore, it may be leveraged for novel nanophotonic devices if suitable materials with high conversion efficiency can be found.

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