Nonlinear optical response of Phosphorene

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Abstract

Few-layer black phosphorus, or phosphorene, has recently attracted substantial attention as a new member of the two-dimensional crystals' family. As a semiconductor with a bandgap in the near infrared region, it bridges the gap between graphene and the large bandgap transition-metal dichalcogenides, with very promising electronic, optoelectronic and photonic applications. Photonics, in particular, is expected to become increasingly important in signal processing, optical computing and quantum information, provided that highly nonlinear optical materials are available to allow for the development of high speed and low energy consumption parametric all-optical devices. However, no characterization of parametric nonlinear optical effects has so far been reported in bulk or few-layer black phosphorus. Here we show that third harmonic generation in phosphorene is highly efficient, with signals being three orders of magnitude larger than with graphene. Furthermore, by monitoring third harmonic generation while black phosphorus is laser thinned, we observed a huge enhancement in efficiency for monolayer samples, in which case the third-order nonlinear susceptibility becomes a hundred of times larger than that of bulk. This enhancement is attributed to the emergence of a doubly resonant process, for a 1550-nm wavelength pump, that is present in the monolayer case, as confirmed by band structure calculations.