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Phase-sensitive Surface Second-harmonic Generation of Topological Dirac Semimetal

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Topological semimetals are a new class of quantum materials characterized by symmetry-protected, nontrivial band crossings near the Fermi energy. These systems have attracted wide interest for their ability to host exotic quantum-transport and electromagnetic phenomena. Although bulk topological semimetals have demonstrated enhanced optical nonlinearities with promise for next-generation optoelectronic applications, intrinsic symmetry constraints often limit the scope of their nonlinear response. In contrast, the topological surface states (TSSs) of these materials have been predicted to support unconventional nonlinear optical signatures under relaxed symmetry conditions. However, surface nonlinear optics of topological materials has remained elusive, and whether it can be enhanced through TSSs is still unclear. In this work, we address this challenge by performing heterodyne-detected second harmonic generation (HD-SHG) spectroscopic study on a prototype Dirac semimetal PdTe₂. We find its reflected SHG to follow *C_{3v}* surface symmetry with a time-varying intensity governed by oxidation kinetics of the material after its surface cleavage, confirming the surface-derived origin of SHG. The complex anisotropic surface nonlinear optical susceptibility tensor χ⁽²⁾ for the pristine surface is fully characterized by heterodyne interferometry, unveiling a hidden giant response along the axis of rotational invariance (|χ⁽²⁾*_{zzz}*|=14±4 nm²/V, corresponding to an effective bulk susceptibility of 14±4 nm/V), where the protected Dirac cones are formed through band inversion. Surface- and orbital-projected band calculations further reveal the correlation of this unusual property to the nontrivial TSSs. Our results highlight the topological interfaces as a promising platform for achieving strong nonlinear optical responses under relaxed symmetry constraints and demonstrate HD-SHG as a powerful *in situ* probe of surface chemistry in topological systems.

Field of Condensed Matter

Topological Materials

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