## High Harmonic Spectroscopy Retrieves Electronic Structure of High-pressure Superconductors

High pressure has revealed surprising physics and creates novel states in condensed matter. Exciting examples include near-roomtemperature superconductivity ( $T_c > 200 \text{ K}$ ) in highlypressured hydrides such as  $H_3S$  and  $LaH_{10}$ .

Although the superconducting transition temperature of high-pressure superconductors is constantly increasing, the mechanism of superconductivity under such high pressure remains an open question. Knowledge regarding the properties and ultrafast dynamics of electrons and quasiparticles in high-pressure quantum states is lacking.

High harmonic generation (HHG) is the upconversion of laser light to radiation carried at multiples of the laser frequency. As the poster child of nonlinear optics, HHG in solids originates from the nonlinear driving of electrons within and between electronic bands by strong-field light-matter interactions. Therefore, HHG spectroscopy naturally contains fingerprints of intrinsic atomic and electronic properties of materials. A lot of excitement emerges around learning about the material's properties through this nonlinear, non-perturbative lasermatter interaction.

With the help of state-of-the-art first-principles time-dependent density-functional theory simulations, Prof. MENG Sheng's group from the Institute of Physics (IOP) at the Chinese Academy of Sciences have studied the ultrafast HHG dynamics in the high-pressure superconductor  $H_3S$ .

The researchers found that HHG in high-pressure superconductors is strongly dependent on the electronic structures and electron-phonon coupling (EPC). Via HHG spectroscopy, they retrieved the band dispersion and EPC, and revealed the significant influence of manybody EPC on electron behavior near the Fermi level. Their results support the phonon-mediated mechanism



The high harmonic generation (HHG) spectra in high-pressure superconductor H<sub>3</sub>S. (Image by IOP)





(a) (b) w/o phono w/ phonon s 0.4 eV 2 0 eV нι н 1600 cm<sup>-1</sup> 10 20 30 40 Energy (eV) 2 3 4 5 6 Energy (eV) w/o phonon w/ phonon (c) (d) 2<sup>nd</sup> VB 0.3 sHHG sHHG 10 sHHG (eV) DFT g<sub>ii</sub> (eV/Å) E (eV) DFPT DFPT (eV 0.0 н н н н

Band structure reconstruction via HHG spectra. (Image by IOP)

based on EPC of high-pressure superconductivity, providing an all-optical approach to probe the band dispersion and EPC of high-pressure quantum states.

This study entitled "Solid-state high harmonic spectroscopy for all-optical band structure probing of high-pressure quantum states" was published in Proceedings of the National Academy of Sciences (PNAS).

Electron-phonon coupling reconstruction via HHG spectra. (Image by IOP)

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