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ICFO sheds light on how superconductivity?s precursor forms

Charge density waves often appear as precursors to exotic quantum phases, such as superconductivity. Understanding how they form in certain materials remains a subject of debate. Now, ICFO researchers and collaborators have studied these charge density waves by applying, for the first time, a laser technique called high-harmonic generation spectroscopy. This new optical method's extreme sensitivity can detect subtle asymmetries in the sample's behavior that eluded earlier techniques. This fundamental knowledge could hold key for the realization of correlated quantum phases (like superconductivity) at room temperature. The technique, reported in Communications Materials, could also be used to study and characterize crystals, 2D materials and nanodevices.

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We usually associate quantum effects with the microscopic realm of atoms, electrons, and photons. Yet some quantum phenomena occur on much larger, even macroscopic, scales. Superconductivity and superfluidity, for instance, allow electric currents or fluids to flow without resistance when ultracold temperatures are reached. Although these exotic quantum phases of matter arise from subtle interactions between charge carriers (such as electrons) and the atomic lattice, their effects manifest in the macroscopic regime.

The **charge density wave** (**CDW**) often appears as a precursor to these correlated quantum phases and could hold the key towards their realization at room temperature. It appears when both the distribution of electrons and the positions of atoms form a repeating, wave-like pattern -a repeating supercell structure. However, the primary mechanism behind CDW formation for some materials (like TiSe2) remains a subject of debate. Some attribute it to interactions between electrons and phonons (collective excitations of the atoms in a solid), while others to correlations between excitons (bound states between an electron that abandons its site in the valence band and the hole it leaves behind). So far, experiments have not clearly distinguished between the two.

ICFO researchers from the Attoscience and Ultrafast Optics group, **Igor Tyulnev**, **Dr. Lenard**

Vamos, **Julita Poborska**, led by **ICREA Prof. Jens Biegert**, along with researchers from the Quantum Optics Theory group, **Dr. Lin Zhang** and **ICREA Prof. Maciej Lewenstein**, have now taken a radically new approach by applying high-harmonic generation spectroscopy (a laser-based technique that makes materials emit light at higher harmonics of the incoming beam) to a CDW system.

The method proved to be extremely sensitive to atomic movement and symmetry changes in the material, revealing that varying atom displacement in different spatial directions affects the CDW strength accordingly. Moreover, with the new ultrafast optical technique the timescale of excitonic response becomes finally accessible, while the process can be modelled via mean-field theory. The study, published in Communications Materials, also involved contributions from ETH Zurich, Adam Mickiewicz University, the Donostia International Physics Center, and the University of Valencia.

Toward solving a long-standing debate around CDW formation

In particular, the team investigated TiSe2 by sending a mid-infrared laser pulse to it and measuring the strength of the resulting harmonics in all directions. The sample was placed in a carefully tailored cryostat chamber, so that the researchers could decrease the temperature down to 14 Kelvin -way below the phase transition temperature of 200 Kelvin, allowing them to access the CDW behavior.

The measurements demonstrated that when atoms shift differently along various directions, the high-harmonic response changes accordingly in each direction. i¿½Even a ti y displacement can trigger a macroscopic change, leading to a very strong variation in t e harmonic signal,i¿½ shares Igor Tyulnev, first author of the article. i¿½This had no been observed with traditional techniques like ARPES, and was only recently hinted at n STM studies -likely because earlier methods dismissed the signal as noise or weren't sen itive enough to detect it.i¿½ As CDW strength is directly tied to atomic movement, the armonic signal effectively served as a sensitive probe of this quantu phase. To better understand the connection between high-harmonic generation atomic displacement, and CDW behavior, the team collaborated with ICREA Prof. Maciej Le enstein and his group at ICFO. Their theoretical insights were crucial for interpre ing the experimental results and uncovering the underlying mechanisms t play. Beyond its scientific insights, the technique also offers practical advantages. As a fully optical and non-invasive probe, it is simpler and more accessible than traditional ethods, which makes it ideal to study and characterize crystals, 2D materials and nano evices. Moreover, since both excitonic and phononic effects influence high-harmonic gen ration, this tool could be used to study them -and potentially distinguish one from th other. According to ICREA Prof. at ICFO Jens Biegert, senior author of the study: i¿½**Our approach could help resolve the long-standing debate about the main mechanism behind CDW formation in TiSe2.** Hopefully, we'll soon be able to answer the question: is CDW driven by electron-phonon or excitonic effects?ï¿

Reference:
Tyulnev, I., Zhang, L., Vamos, L. et al. High harmonic spectroscopy reveals anisotropy of the charge-density-wave phase transition in TiSe2. Commun Mater **6**, 152 (2025).
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TiSe2 sample inside the cryostat.
Credit: ICFO.