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After publication story

Ultrafast nano-movie of graphene

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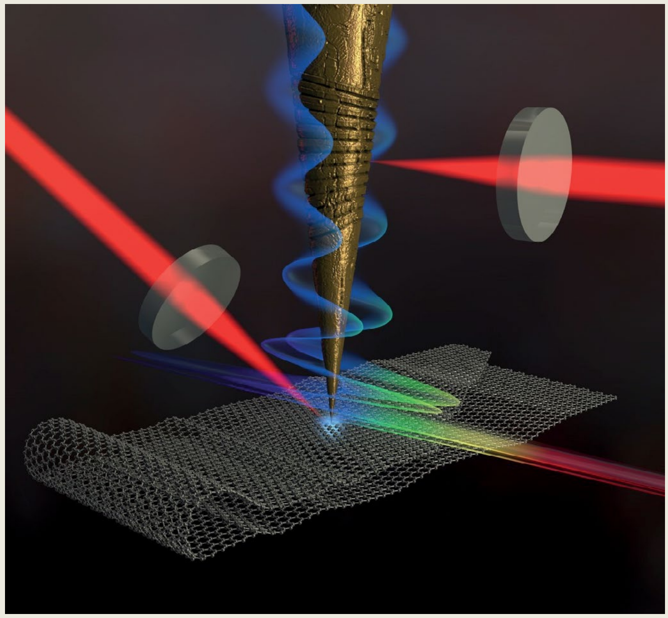
Capturing electron dynamics in condensed matter on a femtosecond timescale means following their motion at the most elementary level. These are processes that govern ubiquitous phenomena including chemical reactions and charge-carrier transport in materials and devices (electrochemical and photovoltaics). In graphene, coherent electron dynamics crucially affects the nonlinear optical response from the terahertz to the ultraviolet, thereby underpinning the working of optical devices for nonlinear frequency converters, ultrafast switches, modulators, and graphene-based information processing units. However, the journey towards fully grasping graphene's unique nonlinear response and the ultrafast electronic coherence has been overshadowed by questions surrounding the role of heterogeneities like defects, edges, and grain boundaries.



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In a paper published in 2019, Jiang et al. implemented an approach to ultrafast nanospectroscopy of 2D materials using nanofocusing of femtosecond pulses on a plasmonic waveguide scanning probe tip. In broadband four-wave mixing (FWM) this allowed for imaging the nonlinear response and coherent electron dynamics in graphene with simultaneous nanometre spatial and femtosecond temporal resolution (T. Jiang et al. *Nat. Nanotechnol.* **14**, 838–843; 2019). A key measurement was the electron decoherence times T_2 ~ 5–6 fs, which highlights an efficient carrier-carrier scattering in graphene.

According to Tao Jiang, the first author of the work, and now at Tongji University in Shanghai: "to actually see a nonlinear response from just a few thousand atoms highlights the exceptional nonlinear optical properties of graphene [and 2D materials in general]."

Due to the high spatial resolution of their technique, Jiang et al. discovered a distinct enhancement of the FWM signal at the edges of the graphene sheets. This observation generated a lot of attention as one of the as yet few direct observations of the nonlocality of a nonlinear response emerging in strong nanoconfined light–matter interaction. "Our theory collaborator, Alexey Belyanin from Texas A&M, attributed this observation to a Doppler effect controlling the nonlinear interaction between the tip plasmons with large near-field momenta and the graphene electrons with their high Fermi velocity," recalls Jiang.

These nonlinear interactions in nanoconfined near-field geometries "opened a regime for studying nonlinear nano-optics effects with new selection rules or enhanced frequency conversion efficiency," reflects Markus Raschke from the University of Colorado, Boulder and principal investigator of the work. Since the initial observation back in 2016 with plasmonic gold hotspots (V. Kravtsov et al. *Nat. Nanotechnol.* **11**, 459–464; 2016), the technique of ultrafast coherent nonlinear nano-imaging has now proven its capabilities to probe the nonlinear optical responses in both graphene (T. Jiang et al. *Nat. Nanotechnol.* **14**, 838–843; 2019) and WSe_2 (W. Luo et al. *Nano Lett.* **23**, 1767–1773; 2023) with nanometre and femtosecond spatiotemporal resolution.

This work shows clear opportunities for nonlinear and quantum photonic devices based on 2D materials. With the virtually infinite potential of 2D-material heterostructures, from engineered novel quantum states to tailored photonic, electronic, and phononic devices, Jiang et al.'s technique could serve as an important probe to investigate the underlying intra- and interlayer carrier dynamics devices.

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