Imaging and Controlling Coherent Phonon Wave Packets in Single Graphene Nanoribbons

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The motion of atoms is at the heart of any chemical or structural transformation in molecules and materials. Upon activation of this motion by an external source, several (usually many) vibrational modes can be coherently coupled, thus facilitating the chemical or structural phase transformation. These coherent dynamics occur on the ultrafast time scale, as revealed, e.g., by nonlocal ultrafast vibrational spectroscopic measurements in bulk molecular ensembles and solids. Tracking and controlling vibrational coherences locally at the atomic and molecular scales is, however, much more challenging and in fact has remained elusive so far. Here, we demonstrate that the vibrational coherences induced by broadband laser pulses on a single graphene nanoribbon (GNR) can be probed by femtosecond coherent anti-Stokes Raman spectroscopy (CARS) when performed in a scanning tunnelling microscope (STM) [1]. In addition to determining dephasing (~ 440 fs) and population decay times (~1.8 ps) of the generated phonon wave packets, we are able to track and control the corresponding quantum coherences, which we show to evolve on time scales as short as ~ 70 fs. We demonstrate that a two-dimensional frequency correlation spectrum unequivocally reveals the quantum couplings between different phonon modes in the GNR.

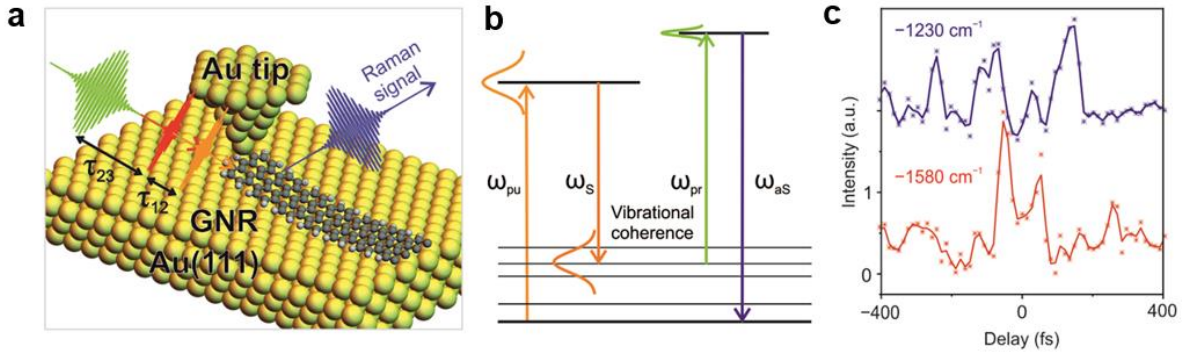

Fig. 1. a, Schematic illustration of time-resolved CARS of a GNR. b, Schematic illustration of impulsive excitation of a vibrational wavepacket in a GNR interacting with ultrashort broadband pump and Stokes pulses. c, Temporal variation of the spectral intensity of two different phonon modes.

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