Probing the optical dipole transition and vibrational coherences in individual (5,4) carbon nanotubes by femtosecond pulse shaping

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Optical detection and coherent control of quantum interferences in nanostructures have attracted considerable attention both as tools to study ultrafast photo-induced phenomena and for their possible application in novel nanophotonic devices. Here, we perform experiments on individual semiconducting single-walled carbon nanotubes (SWCNTs) using femtosecond laser pulse shaping microscopy. Single (5,4) SWCNTs [1] excited with a pair of phase-locked 20 fs laser pulses [2] exhibit beating patterns in their photoluminescence intensity resembling Rabi oscillations in a two-level system. We show that these oscillations can be controlled within the first approximately 100 fs by varying the delay of the pulses and by tuning their relative carrier envelope phase.

Whereas the observed signal variations can be interpreted in terms of coherent control [4-6], they could also be understood in terms of a spectral interference effect because the experiments were carried out mainly in the linear response regime [3]. On the basis of our measurements we discuss the limits of the coherent control approach in pulse shaping microscopy.

For longer pulse separations, coherent radial breathing mode excitations with a period of 90 fs are observed, matching the well-known radial breathing mode in these nanotubes. We show that the onset of these nanoscopic oscillations is deterministic for the (5,4) nanotubes.

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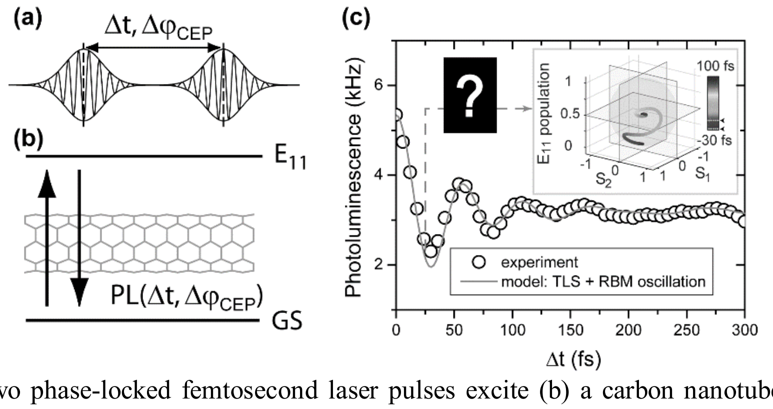

Fig. 1 (a) Two phase-locked femtosecond laser pulses excite (b) a carbon nanotube resonantly. (c) Autocorrelation scans show distinct beating patterns which can be explained in the context of coherent control and Rabi oscillations, but this interpretation depends on the exact circumstances in a critical fashion.