DYNAMICS AT THE NANOSCALE: ULTRAFAST EXCITON PROCESSES IN SINGLE WALL CARBON NANOTUBES

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The extreme aspect ratio of single wall carbon nanotubes makes them perfect candidates for investigating ultrafast exciton processes in the 1D confined space. A rich phenomenology is appearing, which is the ground for rationalize nanotubes photonic properties and possibly better exploit them. We report on results obtained by applying a variety of ultrafast non-linear optical spectroscopies, such as pump-probe, pump-push-probe, coherent phonons and saturation spectroscopy to carbon nanotubes ensembles in solid matrix. Photo-excitation dynamics is studied in chiral enriched samples which contain a large fraction of (6,5) tubes. We use sub-20 fs optical pulses and cw excitation for probing short and long lived states. The sizable reduction in homogenous broadening allows extracting a well defined scenario of energy relaxation for semiconducting single wall carbon nanotubes (CNT). The excitation with sub-20 fs pulses in resonance with the first exciton transition provides a way for determining the exciton size in (6,5) CNT by exploiting the phase space filling model. Pump intensity dependent measurements of the nascent photobleaching show saturation linear with pump intensity, according to the simple saturation law predicted by the model and suggest a characteristic length of the exciton envelope wavefunction in the order of 10 nm. Pumping both first and second exciton resonances we get information onto the initial dynamics, which points to the several processes occurring in competition, namely non radiative exciton deactivation and exciton-exciton interaction (Auger-like process), followed by second-exciton regeneration and ultrafast internal conversion back to first exciton. Time scale and characteristics of all processes are obtained. Impulsive excitation induces coherent phonons in both inhomogeneous and more homogeneous films. Wavelength dependence and phase relationship are investigated and quantum mechanical modelling is carried out in order to rationalize the finding. Anharmonicity of phonon dynamics, leading to non adiabatic mode coupling is directly detected in the time domain. CW photomodulation spectroscopy reveals long lived states are formed in CNT samples, possibly associated to trapped charges which induced the electro-optic effect.