ULTRAFAST RELAXATION PATH DUE TO COLLIDING COHERENT EXCITONS IN THE STRONG COUPLING REGIME

Gilliot, Pierre[1*], Gallart, Mathieu[1], Hönerlage, Bernd[1]

[1] *IPCMS, CNRS and Université de Strasbourg, Strasbourg, France*
**e-mail: pierre.gilliot@ipcms.unistra.fr*

Exciton-exciton annihilation is a very essential process that governs excitation dynamics in photo-physics. For example, the efficiency of carrier generation in photovoltaic organic materials is limited by exciton-exciton annihilation that decreases their population before they have been dissociated. As well, the light-emission efficiency of carbon nanotubes is strongly reduced due to the relaxation that follows collisions between excitons [1]. Moreover these processes seem to be very efficient and very fast: An instantaneous E_{22}- to E_{11}-exciton transfer is observed in carbon nanotubes [2] and the measured very short relaxation times are incompatible with the usual processes, involving for example a cascade of phonons that would imply a minimum relaxation time for each phonon emission [3]. Furthermore, when exciting the lowest E_{11}-exciton resonance, an instantaneous change on the E_{22} resonance [4] is observed, showing a direct link between the two states.

Exciton-exciton annihilation is usually seen as an irreversible process that resembles an Auger recombination: the collision between two excitons gives rise to an annihilation of one of them, while the other one is sent to a high energy level. We show, in the present work, that, instead of being connected by a one-way irreversible process, the two-exciton state and the high-energy one-exciton state can be in a strong-coupling regime. This induces a coherent superposition of these two states that can persist up to the relaxation of one of the two components.
We describe the two-exciton states using the Schwinger representation of su(1,1) algebra as eigenstates of a pseudo-angular momentum [5]. This two-exciton manifold is coupled to the higher energy one-exciton states. In the case of carbon nanotubes for example it is enhanced by the resonance between twice the energy of the E_{11} exciton and the energy of the E_{22} exciton. Together with the high strength of the interaction, this gives then rise to a strong coupling the two manifolds.

Strong coupling has deep consequences on the exciton dynamics. First the exciton population can be coherently transferred to the high energy states in a very efficient way. Second, as in a degenerate parametric amplification process that is described by the same Hamiltonian, at high exciton densities, the E_{11} population is depleted in favor of the E_{22} level. The latter being less protected from the interaction with the environment, this favors non-radiative exciton recombination. We will discuss in this paper the efficiency of this process and relate it to the spatial features of the excitons described by their diffusion and coherence length [6] that govern the collision efficiency.

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