Efficient exciton energy recycling in Si nanocrystals

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A nonradiative Auger recombination of an exciton and an impact excitation by a highly energetic free carrier are two inverse manifestations of the same physical process. Two excitons may undergo Auger recombination, forming a single, high-energy electron-hole pair, while one of the "hot" carriers created in this process can in turn, induce a band-to-band excitation, thus reproducing the lost exciton and reversing the Auger process. The sequential action of these events will continue as long as the excess energy of a hot carrier will be sufficient to induce the impact excitation. Therefore the duration of this specific "energy recycling" will be controlled by the hot carrier cooling processes. Commonly the most important of these is the phonon scattering. Since this is process is very efficient in bulk semiconductors, the energy recycling is not an important phenomenon for these materials.

The situation is different for semiconductor nanocrystals where quantum confinement has a profound influence on all the processes relevant to energy recycling. The enhanced Coulomb interaction dramatically increases Auger recombination and impact excitation rates, while level discretization may restrict electron-phonon interactions, slowing down the cooling. Consequently, the energy recycling process gains in importance. This can lead to enhancement of effective populations of some specific higher energy states, prolongation of the effective lifetime of multiple excitons localized within the same nanostructure, new phenomena of photon up- and down-conversion, and stronger coupling between individual nanocrystals, among others. The efficient Auger energy recycling has been proposed as being responsible for several phenomena of enhanced energy transfer on nanoscale, and in particular for the effective sensitization of rare-earth emitters and for the exciton exchange between proximal nanocrystals.

In my presentation, I will discuss energy recycling in dense ensembles of Si nanocrystals. I will introduce the concept and then present experimental support for the existence of this phenomenon. I will argue that the efficient process of exciton energy recycling is responsible for the following manifestations:

- Enhanced hot carrier emission: Upon intense pumping, the increase population of high energy electron states enhances the radiative recombination of the phoneless transition in the Γ-point of the Brillouin zone (the "direct bandgap" recombination)
- Emission fingerprint of the multiple exciton generation: In dense dispersions of Si nanocrystals in SiO_2 matrix prolonged recycling of the hot carrier state allows to effectively transfer the excess energy to a neighboring "unexcited" nanocrystal, thus arresting the Auger process and enabling radiative recombination of the second exciton
- Enhanced emissivity of bi-exciton: Due to efficient recycling the effective lifetime of the bi-exciton state is prolonged, resulting in additional luminescence, above the saturation level determined by the Auger process between excitons co-localized in the same Si nanocrystal.

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