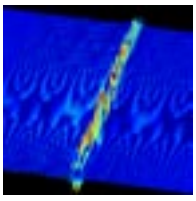
Ultrafast Spectroscopy of Nanostructures



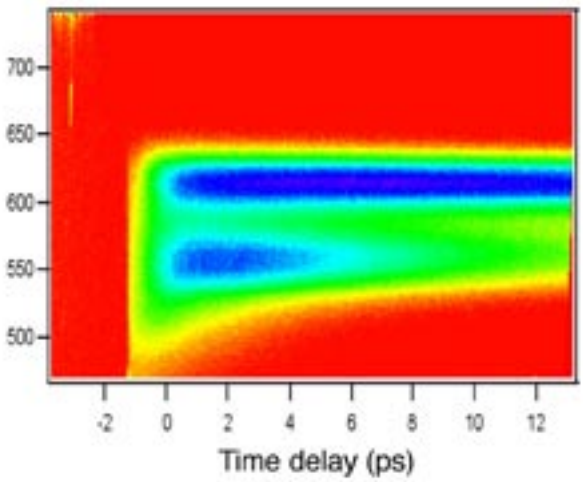
Nanostructures and nanoparticles are systems, which can serve as a basis for a very wide range of photonic devices. A proper understanding of the interaction of light with such objects is a prerequisite for most of these possible applications. In this project, we use ultrafast spectroscopic tools to probe the interaction between light and different types of nanostructures.

Chemically prepared nanostructures.

The ultrafast relaxation dynamics of charge carriers in semiconductor nanostructures prepared by chemical methods is the main focus of this research. This project involves improving upon the growth of nanoparticles, with a size ranging between 1 and 10 nm, and studying them by femtosecond fluorescence and transient absorption techniques.

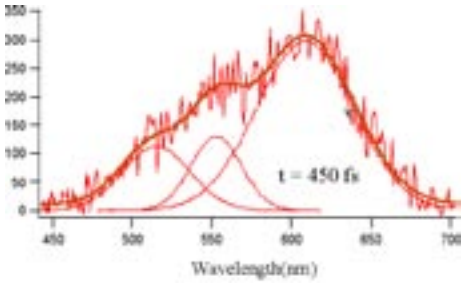
The chemical synthesis has been improved by using both organic and inorganic ligands. This allows for flexibility in the choice of applications for our nanoparticles. In particular, capping them with hexadecylamine (HDA) has allowed us to improve the variation in size distribution to less than 5% and to obtain fluorescence quantum efficiencies of ~30%.

A femtosecond (fs) fluorescence up-conversion set-up



Spectrogram (wavelength versus delay plot) of 3.2 nm CdSe nanodots fluorescence (vertical axis in nm, horizontal in ps)

has been built that combines a high temporal resolution of 50-70 fs with the spectral analysis of the fluorescence from 480 to 730 nanometers (nm). Using this set up we can probe the dynamics of the electron population in the different levels of the dot by following the evolution of the luminescence very precisely. Over the last year, we have studied isolated nanoparticles of CdSe, in the form of dots and rods, to identify the effect of laser power, capping material and the solvent used.



Transient emission spectra of 3.2 nm CdSe nanodots for a time delay 450 fs

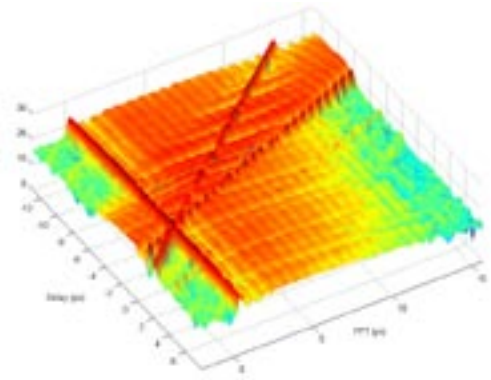
Fluorescence up-conversion measurements on dots enable us to not only capture the time profile of the band gap fluorescence but to also follow the emission from intermediate steps in the relaxation. As an example, the transient fluorescence spectrum at 450 fs for 3.2 nm dots shows that we evidence a three-component emission band. The kinetics of these bands have been systematically recorded as a function of excitation density, and we observe unexpected band filling effects. Similar qualitative features have been observed in the case of nanorods.

Semiconductor nanostructures

Probing the coherence properties of semiconductor nanostructures after interaction with light is the main theme of the work in this area. We have developed different experimental tools that allow us to access such coherence properties.

One of these techniques consists in probing the interference pattern of the light emitted by a given sample, for example a semiconductor quantum well, with the laser beam used to excite the sample. Using this technique, called spectral interferometry, we can retrieve the dynamics of the coherence through a Fourier transformation of the observed spectrum. In the figure shown here, the coherent signal from GaAs quantum wells is reported.

Nanoscale optics



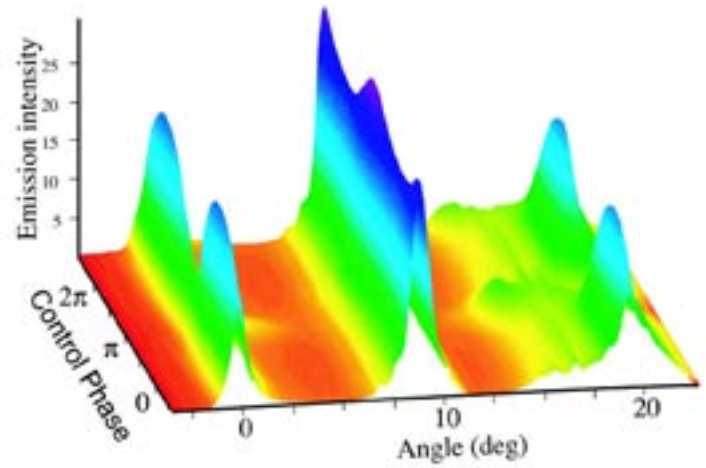
Fourier transform of the spectral interferogram, showing the coherent Rayleigh signal emitted by a quantum well after excitation by a femtosecond pulse.

Quantum beating of the heavy hole and light hole excitons is used to extract the coherence time between these two excitations of the system.

The system on which we have concentrated most of our attention thus far, is the parametric amplification of polaritons in microcavities. In microcavities, the light field is strongly modified by the two mirrors forming the cavity. This results in a very strong interaction between light and the quantum wells placed in the cavity. In such a case, the excitations of the system are called polaritons. These particle possess very interesting properties. One of them is that they can strongly amplify a light beam, with an ultrafast time response. The states giving rise to this amplification process must have, according to theories, very peculiar coherence properties.

The coherence properties of the system of polaritons in microcavities can be probed using the technique called coherent control. In this experiment, two probes are amplified in the system, but they keep a very precise phase relation, which allows the second probe, to destroy the amplification of the first one, even after delays much longer than the duration of any of the two probe pulses.

We have evidence that the amplication can be so controlled, especially at normal incidence (0°), where we monitor the probe amplification as a function of the phase between the two probes. At the angle of 10°, the pump appears to be depleted, when the probe beam is amplified, and replenished when the probe is deamplified. These measurements demonstrate the coherence of the polariton population in the microcavity.



Angularly resolved emission of the microcavity, as a function of the phase between the two control pulses, evidencing control over the entire emission pattern, including the idler. The delay is here 2 ps, much larger than the width of the two pulses.

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