Coherence-time measurements of single photons emitted by CdSe colloidal nanocrystals

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CdSe nanocrystals are often studied as prototypical colloidal quantum dots. They can be easily manipulated and exhibit many promising optical properties, like a broad absorption spectrum and a sharp emission line. Their emission wavelength can also be tuned between 450 et 600 nm by controlling the nanocrystals synthesis parameters. They have found a wide range of applications from optoelectronic devices to biological labelling or imaging. In the field of quantum optics, triggered single photon emission under pulsed excitation has been demonstrated recently [1].

The realization of a single photon source is of great interest for quantum cryptography. In this case, the coherence of the single photons emitted is not required. However, single photons could also be used for quantum information processing. A recent proposal is based on the interferences between various photons. For such an application, the coherence of the single photons becomes crucial. One idea to obtain coherent photons is to use single quantum systems the emission linewidth of which is not enlarged by dephasing mechanism. The dipole coherence time is then equal to twice the radiative lifetime. Interferometric studies for single self-assembled InAs/GaAs quantum dots at low temperature have shown decoherence times as long as 650 ps, and two-photon interferences have been demonstrated.

Spectroscopic studies of single CdSe nanocrystals have shown the diffusion of the wavelength emission over a few meV, related to changes in the electronic environment [1]. At low temperature, the emission spectra measured by standard spectroscopy or interferometry should be sharper than 0.2 meV but they are broadened by this spectral diffusion. Faster spectroscopic measurements are not possible because of weak emission intensity (at most 20.000 counts/s collected at low temperature).

In this paper, we first present an original method which we call photon correlation Fourier spectroscopy (PCFS), which allows us to achieve a high resolution in both the temporal and spectral domain, and measure the emission coherence in spite of weak emission and fast spectral diffusion. It is based on the time-correlation between the photons detected at the outputs of a Michelson interferometer. We explain how this method allows one to measure the coherence time of a single emitter fluorescence despite wavelength fluctuations at short time scale. We investigate theoretically the behavior of intensity correlations at very short timescales, and determine general expressions for the spectral resolution of PCFS. For a spectral diffusion of 25 ŏ3bceV on a 0.1 ms timescale, as reported in the literature [2], we predict a resolution of a few ŏ3bceV, three orders of magnitude better than standard spectroscopic or interferometric measurements.

This method is particularly fitted for the study of CdSe nanocrystals. We present the precise measurement of the time coherence of the CdSe QD fluorescence. A previous spectroscopic study of single CdSe nanocrystals has shown the diffusion of the wavelength emission over a few meV, related to changes in the electronic environment [5]. At low temperature, where emission linewidth are expected to be below 0.2 meV, direct linewidth measurements are not possible, due to insufficient spectrometer resolution and spectral diffusion during the duration of the measurement.

The experimental setup consists in a standard confocal microscope and a Michelson interferometer. By proper normalization, we obtain the Fourier transform of the emission spectrum broadened on a time ŏ3c4, yielding information (1) on spectral diffusion and (2) on the emission linewidth (by taking ŏ3c4 below the spectral diffusion timescale).

Interferometric measurements at room temperature have led to linewidth values around 130 meV. At

4 K, first measurements show typical linewidths of around 3 meV. This high value may result from non-resonant excitation. We also present the temperature dependance of the coherence time and the lineshape study.

References

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