



# GEMaC

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## CDSE/ZNS NANOCRYSTALS

In the case of CdSe/ZnS nanocrystals, the fluorescence is characterized by "dark periods" corresponding to blinking phenomenon. The duration of these periods follows a statistical law called Lévy distribution which is a power law without mean or standard deviation. A key feature is the relatively frequent occurrence of long events.

This property has fundamental consequences such as non-ergodicity or "aging statistics" of the fluorescence of a set of nanocrystals [Brokmann et al., Phys. Rev. Lett. **90**, 120601, 2003]. To characterize the nanocrystal as a single photon source, we studied the collection, as high as 7%, of emitted photons [Brokmann et al., Appl. Phys. Lett. **85**, 712, 2004]. The collection of single photons has been studied through the effect of a dielectric interface on the fluorescence nanocrystals: modifications of the radiation pattern of the nanocrystal; modifications of the radiative lifetime [Brokmann et al., Chem. Phys. Lett. **406**, 210, 2005, Brokmann et al., Chem. Phys. **318**, 91, 2005]. A defocused imaging method was used to determine the orientation of the nanocrystal in space. This study also allowed us to determine the radiative quantum efficiency of a nanocrystal at the single emitter level [Brokmann et al., Phys. Rev. Lett. **93**, 107403, 2004]. This data, essential for the characterization of the optical properties of these nanoparticles, appears close to unity. All this work has finally shown that nanocrystals represented very promising single photon emitters

Other studies have focused on the temporal coherence of the nanocrystals. Indeed, a recent proposal to achieve optical quantum logic gates suggests to take advantage of a phenomenon known as coalescence between two single photons: when two identical photons are simultaneously incident on both input channels of a beam splitter, they always get out by the same output channel. The quantum nature of the interference can be observed from single photons spectrally monomode. These photons are emitted by a single dipole when the transition is not broadened by any dephasing mechanism: the coherence time of the dipole  $T_{c2}$  then reaches its upper limit value, equal to twice the radiative lifetime  $T_{r1}$ . However, if the dipole is subject to fluctuations, the photons will follow these fluctuations which vary in time and they will be discernible. Until recently, the temporal coherence of a nanocrystal was unknown. In fact, the emission wavelength of a nanocrystal varies by jumps (this phenomenon is called spectral diffusion) at short time scales, below the integration time of a typical standard spectroscopy experiment (100 ms). Then the linewidth measured corresponds to the amplitude of the spectral distribution during the acquisition period. An original technique of Photon-Correlations Fourier Spectroscopy (PCFS) [Brokmann *et al.*, Opt. Exp. **14**, 6333, 2006 - Coolen *et al.*, Phys. Rev. A **76**, 033824, 2007] was used to characterize the dynamics of spectral diffusion at short times and to more accurately assess the line-width of the emission partially independent of the broadening effect due to the spectral diffusion. The measurements were used to characterize the dynamics of spectral diffusion of a nanocrystal of CdSe/ZnS at 10 K and up to 10 microseconds. At this scale, the coherence time measured reaches values as large as 200 ps [Coolen *et al.*, Phys. Rev. Lett. **100**, 027403, 2008], which is a better value than the 1 ps time obtained by conventional spectral measurements. These measurements also showed that the spectral diffusion was still active for delays as long as 10 microseconds.