Phonon impact on a single CdSe quantum dot from cryogenic to room-temperature

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An excellent way to produce single photons is to use solid-state emitters, particularly quantum dots (QDs). They are promising single-photon sources due to their ability to emit bright, pure, and indistinguishable single photons on demand through the radiative cascade of excitonic states (e.g., the neutral bi-exciton, exciton and charged excitons). High purity is achievable only when the collected photons come from a single spectral line, typically obtained at cryogenic temperature where the emission lines are narrow. As the temperature increases, the lines broaden, and the mixing between them diminishes the purity. Thus, it is essential to understand the broadening mechanisms that degrade the performances of the source at non-cryogenic temperatures.

In this contribution, we study the linewidth broadening of a CdSe quantum dots embedded within a ZnSe nanowire (inset of Fig.1), which is a promising system for room temperature single photon emission in the blue-green range, a spectral window of interest for free-space quantum communications [1]. Unlike Stranki-Krastanov QDs, where a wetting layer surrounds the QD, our emitter is inserted within a material of higher bandgap, thus preventing the escape of charge carriers at higher temperatures. In addition, the large X-XX splitting (22.4 meV) observed in our system allows us to distinguish the different lines composing the spectrum at elevated temperatures and, therefore, is particularly suited for studying the temperature-dependence of the emission of a single CdSe QD.

At 6K, each emission line comprises a narrow zero-phonon line and a phonon sideband due to the interaction between a localized exciton and longitudinal acoustic phonons [2]. We present an extension of the Huang-Rhys theory to the exciton-phonon interaction within a single QD, yielding a convenient analytical expression capable of describing the significant sideband broadening with temperature, to the detriment of the zero-phonon line (Fig. 1). The model accounts for multi-phonon interactions via the deformation potential and fully captures the temperature dependence of the spectra up to 300K. A set of parameters can be inferred from low-temperature spectra and are used to fit accurately higher temperatures, as shown in Fig. 1. The ratio of the two components of the phonon sidebands, is a good measure of the local temperature.

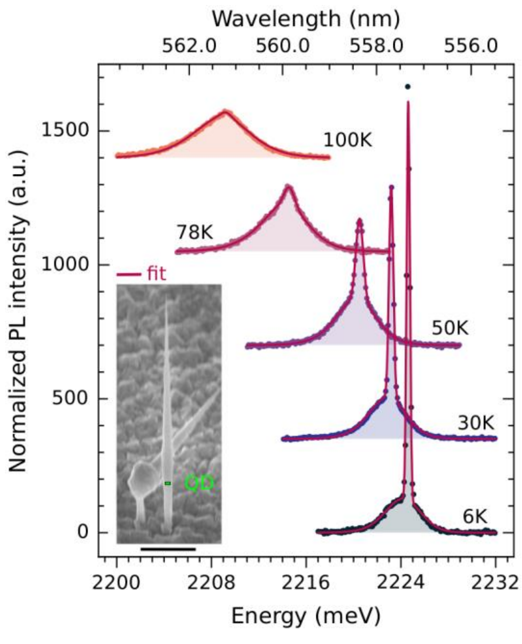

Fig.1. Temperature dependence of the exciton line. The dots and the solid line represent the experimental photoluminescence data and the model respectively. The studied emitter is shown in the inset (scale bar 1µm).

Additional lines, such as charged exciton lines, must be considered depending on the nanowire. A significantly larger Huang-Rhys factor *S* is observed for the charged exciton than the exciton. Above 100K, both neutral and charged (bi) excitons partly overlap, highlighting the need for an efficient control of the charges to take full benefit from the large binding energy of the neutral bi-exciton. Ultimately, the model explores the mechanisms degrading the purity of the source and determines the operational temperatures with good purity and brightness. This work paves the way toward implementing QD-based sources operational at non-cryogenic temperatures via a Peltier cooler.

References

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