Phd Thesis Defense: Optical Spectroscopy of Infrared--Emitting Colloidal Quantum Dots: From Ensemble to Single Particles

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Colloidal quantum dots (CQDs) have established themselves as a key technology in optoelectronics due to their high efficiency, versatility, and low manufacturing costs. Their application in light generation has evolved from statistical thermal emission in visible LEDs and fluorescence converters toward more sophisticated sources, such as lasers and quantum light emitters. However, their implementation in the infrared (IR) has proven more complex due to an inherently low photoluminescence quantum yield (PLQY), with efficient applications emerging only recently. Given the growing interest in quantum technologies, single photons in the infrared telecommunications C-band have become a fundamental pillar,

a domain where these colloidal technologies offer superior flexibility. Specfifically, CQDs based on Pb chalcogenides and III-V semiconductor alloys bridge the gap between cost and integration thanks to the tunability of their emission properties within this spectral region. This work presents an optical spectroscopy characterization of PbS and InSb CQD dispersions and films, employing steady-state and time-resolved micro-photoluminescence (u-PL) techniques at low temperatures. To this end, the complete design and implementation of the utilized optical system are detailed, including alignment and optimization protocols, as well as the algorithms developed for performing confocal u-PL mapping with micrometric resolution. Power-dependent PL studies on PbS films reveal a collective behavior characterized by a bimolecular recombination regime within a band with an exponential-type density of states (DOS), which contrasts with the excitonic behavior observed in solution. Furthermore, the PLQY of these samples shows a marked linear dependence on size, reaching a maximum of ~30\% for quantum dots with excitonic peaks around 1.5 eV and steadily decreasing toward larger particles. Time-resolved PL measurements indicate the existence of energy transfer mechanisms between CQDs with long diffusion times. Encapsulation tests confirm that oxygen acts as a highly degrading agent, quenching emission after moderate exposure to ambient conditions. InSb CQDs exhibit extreme sensitivity to oxidation, losing their PL within seconds upon contact with air. Temperature-dependent PL measurements further reveal the presence of a surface-associated emission band at cryogenic temperatures, which rapidly disappears upon heating the sample or adding a thin InP shell. Two strategies were explored to reach the single-particle regime. In contrast to the common use of spin-coating, dip-coating of dilute solutions proved effective for obtaining isolated CQDs with controlled areal density, albeit at the cost of significant luminescence degradation; this suggests irreversible damage to the crystal surface due to ligand stripping subsequently oxidating the surface. As an alternative for protecting particles from external agents, PbS CQDs were encapsulated in thick silica shells using a microemulsion method in combination with surface chemistry engineering, succesfully allowing a high yield of single CQD to silica particle. The results presented in this thesis highlight critical limitations in the current implementation of IR-emitting CQDs as quantum light sources, underlining the need to develop new synthesis routes and coating chemistries to improve their optical properties and ensure long-term stability. Additionally, the experiments conducted on films provide relevant perspectives for the optimization of technologies based on infrared CQDs, such as LEDs, photodetectors, or lasers.

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