

Optical Gain and Stimulated Emission in Nanocrystal Quantum Dots

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Semiconductor quantum dots (QDs) promise the lowest lasing threshold for semiconductor media. Additionally, QDs in the strong confinement regime have an emission wavelength that is a pronounced function of size, adding the advantage of continuous spectral tunability simply by changing the dot radius. Lasing has previously been demonstrated for epitaxially grown III-V QDs [1]. Large lateral dimensions and difficulties in size control limit their spectral tunability using quantum confinement effects. An alternative approach to fabricating QDs is through chemical synthesis which can produce semiconductor nanoparticles (nanocrystal QDs) with radii from 1 to 6 nm and with size dispersions as small as 5% [2]. Such dots show strong quantum confinement and permit size-controlled spectral tunability over an energy range as wide as 1 eV. The combination of tunable electronic energies and chemical flexibility make colloidal QDs ideal building blocks for the bottom-up assembly of optical device structures, including optical amplifiers and lasers.

However, despite more than a decade of effort, lasing in small-size colloidal nanoparticles has not been realized. To determine what hinders lasing action we performed extensive dynamical studies of radiative and nonradiative processes in colloidal QDs. In particular, we demonstrate that nonradiative carrier losses in QDs are dominated not by surface trapping, as was thought initially, but by intrinsic Auger recombination [3]. Auger recombination is a nonradiative multiparticle effect that turns on as soon as two or more electron-hole (e-h) pairs per dot are excited. Since lasing also requires at least two e-h pairs per dot, the Auger effects are intrinsically unavoidable if the condition for lasing is satisfied. Our data indicate a dramatic shortening of the two e-h pair life time (from 363 to 6 ps) with reducing the QD radius from 4.1 to 1.2 nm. However, despite very short time constants, Auger recombination does not inherently prevent lasing. Lasing can still be realized if the development of stimulated emission occurs faster than the Auger decay. The rate of the stimulated emission buildup is proportional to the dot concentration in the sample. We show that by close packing dots into solid state films (QD solids) it is

possible to obtain dot concentrations that are sufficiently high for the optical gain to successfully compete with the Auger decay. In Fig. 1, we demonstrate the development of stimulated emission in a QD film fabricated from CdSe QDs with radius 2.1 nm [4]. This result provides a proof-of-principle for lasing in small-size QDs.

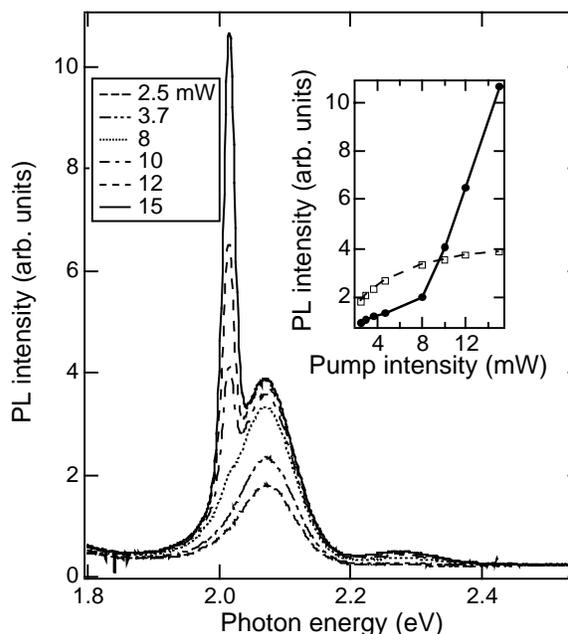


Fig. 2. Development of a sharp stimulated emission band in photoluminescence (PL) spectra of the film fabricated from CdSe QDs with radius 2.1 nm. Inset: Pump dependent PL intensity inside (circles) and outside (squares) the sharp stimulated emission peak.

We also investigate the use of colloidal QDs for realizing lasing in different cavity configurations. In particular, we demonstrate the development of microring lasing modes in the case of QDs deposited on the inner surfaces of capillary tubes.

Our work on optical gain and lasing in colloidal dots can potentially affect many optical technologies by stimulating the development of QD-based lasers and optical amplifiers with operation wavelengths that can be selected over a wide range of spectral energies.

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