Single-mode Tuneable Laser Operation of Hybrid Microcavities based on CdSe/CdS Core/Shell Colloidal Nanorods on Silica Microspheres

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Colloidal core/shell semiconductor nanonorystals have generated a great deal of interest as gain media in recent years due to a number of salient properties originating from their small size and the associated quantum confinement [1]. These include low-threshold and temperature-insensitive lasing, reduced trapping of excited carriers, and the possibility to alleviate non-radiative Auger recombination by engineering the wavefunction distributions of the electrons, and holes within their volume. Here, single-mode, tuneable operation of fiber-coupled hybrid lasers based on colloidal CdSe/CdS core/shell nanorods on silica microspheres is reported.

The nanocrystals were asymmetric, quasi-type-II heterostructures with a 3-nm-diameter spherical optically active CdSe core located at one end of the wider band-gap CdS rod. They were synthesized with the seeded-growth method [2], had an overall size of 28 nm × 4 nm and exhibited a photoluminescence maximum near 610 nm. Microspheres with measured Q factors in excess of 10^8 were produced by rotating an optical fiber taper and simultaneously heating its end with a CO_2 laser beam; they were coated by immersion in a toluene solution of the nanorods. They were pumped at λ~400 nm with a tuneable frequency-doubled Ti:sapphire amplifier, emitting 180-fs-short pulses with a linewidth of 5 nm, at 250 KHz. Pump pulses were evanescently coupled into the spheres with adiabatic tapers made of single-moded fibers at 405 nm. Laser signals were collected by either the same taper used for pumping or, a fiber tip with a diameter of ~50 nm [3]. The pump efficiency was optimized by phase-matching the propagation coefficients between the propagating mode in the taper and the fundamental whispering-gallery-mode (WGM) in the microsphere [3].

Single-WGM operation near 628 nm on the ^1S_h-^1S_e(CdSe) biexciton transition of a 9.2-μm-large hybrid sphere was observed by resonant pumping a fundamental WG pump mode with m = l, where m and l are the angular momentum and the azimuthal numbers, above an absorbed pump power threshold of 67.5 μW (Fig. 1). Such modes are confined to the equatorial ring of the sphere and were identified by a dip observed in the transmission through the taper when the pumping wavelength was tuned [4]. A laser power of 5.5 μW for 155 μW of absorbed power was obtained, corresponding to a slope efficiency of 6.4% (Fig. 2). The laser line had a full-width-half-maximum (FWHM) of Δλ = 0.06 nm, which suggests a Q = (λ / Δλ) value of approximately 10^5 for the hybrid microsphere. Laser emission from spheres with larger diameters was multimode due to the smaller free spectral range (FSR) in-between the modes. Coupling of the pump beam into the resonator away from its equatorial zone results in simultaneous lasing on the biexcitonic (^1S_h-^1S_e), and multiexcitonic (^1P_h-^1P_e) transitions (Fig. 3).

The lasing wavelength was tuned by heating with 3.5-μm-pulses from a tuneable femtosecond laser (200 fs, 80 MHz), which were directed to the microsphere by a 4x objective lens. The laser power was raised in successive steps and for each step the laser spectrum was recorded, indicating a 2.1-nm red-shift of the emission within the available power range (60 mW), which corresponds to 30% of the FSR at the lasing wavelength. The shift observed is due to a temperature-induced increase in the size of the microspheres and changes of the thermo-optic coefficient, band gap, and size of the CdSe/CdS quantum rods.

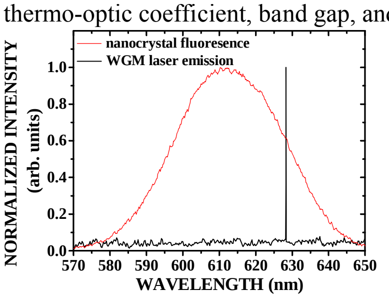

Fig. 1 Single-mode laser emission spectrum of a 9.2-μm-large microsphere

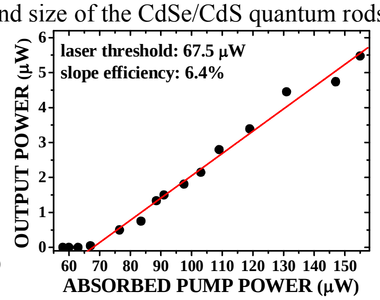

Fig. 2 Power characteristics for single-mode lasing of a 9.2-μm-large sphere.

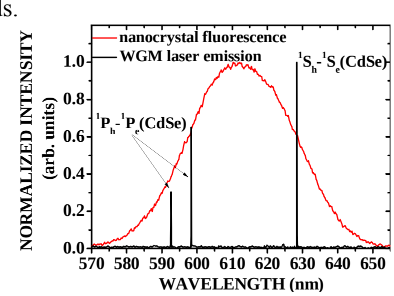

Fig. 3 Laser spectrum for non-equatorial pumping of a sphere with 9.2-μm diameter.

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