CdSe/CdS core-shell heterostructures: Amplified spontaneous emission, gain, lasing and acoustic phonon behavior

M. Miscuglio,[a,b] M.L. Lin,[c] F. di Stasio,[a] A. Polovitsyn,[a,b] I. Angeloni,[a,b] I. Moreels,[a] R. Proietti Zaccaria,[a] G. Manfredi,[b] P. Lova,[b] D. Comoretto,[b] L. Manna,[a] P.H. Tan,[c] and R. Krahne[a]

[a]*Istituto Italiano di Tecnologia, Genoa, Italy*
[b]*Dipartimento di Chimica e Chimica Industriale, Unversità di Genova, Genoa, Italy*
[c]*Institute of Semiconductors, Chinese Academy of Science, Beijing, China*

Colloidal nanocrystals (NCs) are attractive materials for light-emitting applications thanks to their flexible synthesis, size-dependent properties and bright emission. In particular, CdSeCdS core-shell nanocrystals show extremely bright luminescence, demonstrate low-threshold optical gain and even lasing from self-assembled coffee rings [1,2].

We report on broadband amplified spontaneous emission (ASE) from wurtzite CdSe/CdS "giant-shell" nanocrystals (g-NCs) with unprecedented large core up to 7.5 nm in diameter.[3] The combination of large core and shell enable ASE from different CdSe optical transitions as well as from the CdS. Thin films of g-NCs with large CdSe core (7.5 and 5.1 nm in diameter) show ASE at different colors with a similar threshold. Tuning of the core diameter allows to obtain ASE in a wide spectral range, and blending of g-NCs with different core sizes gives rise to a continuous amplified spontaneous emission band from green to red (Fig. 1a). Drop-cast films of such CdSe/CdS g-NCs manifest simultaneous multi-color random lasing under fs- and ns-pulsed excitation (Fig. 1b).

Acoustic lattice vibrational modes in cadmium chalcogenide nanocrystals are closely related to the carrier dynamics of excitons in confined systems and therefore on their optical properties. We studied the acoustic phonon modes in such spherical and dot-in-rod heterostructures by non-resonant Raman spectroscopy and finite elements simulations, and reveal their peculiar behavior that originates from the dot-in-rod architecture (Fig. 1c,d).[4]

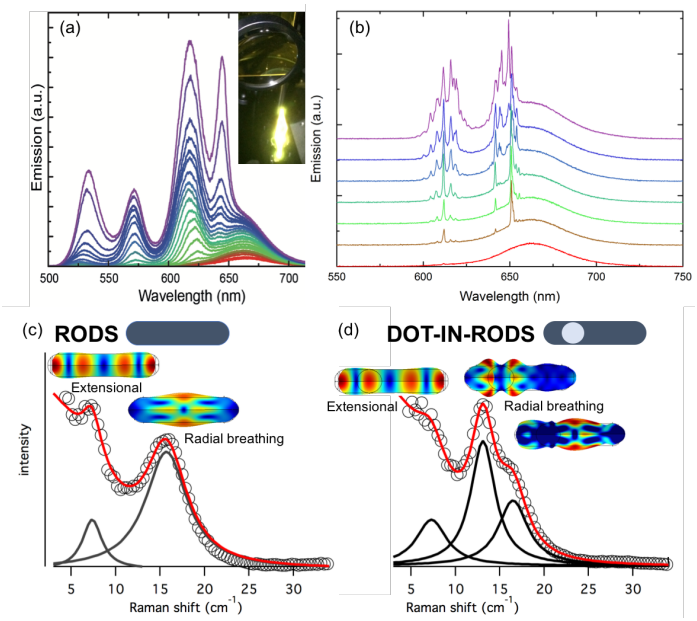

Fig. 1 (a) Broad band ASE (a) and random lasing (b) from films of giant-shell CdSe/CdS nanocrystals under increasing fs-pulsed excitation. (c-d) Radial breathing and extensional vibrational modes of nanorods (c) and dot-in-rods (d) recorded by non-resonant Raman spectroscopy.The insets show the phonon-iduced strain.

Finally, we show a solution-based and bottom-up fabrication process for all-polymer planar microcavity with CdSe/CdS DiRs as emissive medium. In one approach, the DiRs are embedded in a polystyrene matrix to obtain a highly processable nanocomposite, and we discuss the potential of the microcavity for spectral and spatial reshaping of the nanocrystal emission. In a different approach, we achieved single mode lasing by fabricating the solution processed DBRs separately, and by using a pure DiR film as cavity layer.

1) M. Zavelani Rossi et al., *Laser & Photonics Reviews* 6, 678 (2012)
2) F. Di Stasio *et al.*, *Small* 11, 1328 (2015).
3) F. Di Stasio el al., *ACS Photonics* 3, 2083 (2016)
4) M. Miscuglio *et al.*, *Nano Letters* 16, 7664 (2016)