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Thresholdless Optical Gain using Colloidal HgTe Nanocrystals

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Abstract: Thresholdless stimulated emission is observed using nanometer sized colloidal HgTe nanocrystals, in a broad spectral region covering the entire technologically relevant near-infrared spectrum.
OCIS codes: (140.2050) Dye lasers ; (160.4236) Nanomaterials ; (300.6530) Spectroscopy, ultrafast ; (320.7130) Ultrafast processes in condensed matter, including semiconductors

Colloidal semiconductor nanocrystals[1] or quantum dots (QDs) combine size-tuneable atomic-like electronic energy levels with a suitability for solution-based processing, which makes them ideal for numerous optoelectronic applications. Devices relying on optical gain (i.e. stimulated emission) such as lasers or amplifiers could equally benefit from a tunable, low cost and solution processable gain medium, especially in the technologically relevant near-infrared part of the spectrum (1.3-1.55 μm). Although stimulated emission involving the first exciton transition has been demonstrated for a number of QDs[2,3], their use as a gain medium has been limited. This is largely due to the QDs being an effective two-level system, where stimulated emission and absorption involve transitions between the same discrete band edge states. Since these states are degenerate the average number <N> of excitons (X) per QD must exceed at least one to achieve population inversion, requiring multi-X for effective light amplification. Next to this intrinsic drawback of two-level gain media, multi-X gain is problematic due to the fast non-radiative recombination of multi-X through the non-radiative Auger process, capping the gain lifetime to a few tens of picoseconds. Although the resulting high pumping thresholds can be achieved using optical pumping by ultrafast pulsed lasers, this renders electrically pumped lasers based on colloidal QDs elusive.

In this work, using HgTe QDs, we observe optical gain for average exciton densities per nanocrystal <N> as low as 0.005 ! Moreover, a substantial gain coefficient having an extrapolated lifetime corresponding to the single exciton radiative lifetime is observed. We attribute this performance to the presence of shallow traps creating a 4 – level system. To study the potential for HgTe NCs as a gain material, we use ultrafast hyperspectral pump-probe spectroscopy, a powerful tool to analyse the ultrafast carrier dynamics in nanostructures. It provides not only the temporal, but also the spectral evolution of carrier cooling and absorption bleaching. Samples are pumped using ultra-short (200 fs) pulses followed by a broadband probe pulse that monitors the change in absorption of the sample after the photo-excitation event. The quantity measured in a TA experiment is the differential absorbance: $\Delta\alpha = \alpha - \alpha_0$, which can be rewritten as the non-linear absorbance $\alpha = \alpha_0 + \Delta\alpha$, i.e. the absorbance of the sample after photo-excitation. Note that the occurrence of optical gain at a given wavelength is evidenced by a negative non-linear absorbance at that wavelength. In that case, the sample emits more photons than it absorbs, i.e., stimulated emission dominates over absorption.

The occurrence of thresholdless optical gain is confirmed by two independent means, through analysis of the bleach decay kinetics and the fluence threshold for stimulated emission: the long lifetime of the bleach on the timescale of the radiative recombination of the single exciton and the threshold for stimulated emission which lies far below 1 absorbed photon per dot. The thresholds measured (see figure 1, right) are clearly tending to zero and are limited by residual absorption such as intra-band absorption at longer wavelengths.

The question remains whether the stimulated emission we observe is enough to make realistic feedback structures turn to lasers, i.e., whether the modal threshold gain required for such cavities can be provided by HgTe thin films. The answer lies in the calculation of the intrinsic material gain, i.e., what gain (in 1/cm) could be expected for a fictious material of 100% volume-fraction HgTe nanocrystals. The result is shown in 2 for 2 different nanocrystal sizes (3.1 and 3.6 nm). The actual gain must be corrected for the smaller volume fraction in a close packed layer (f =

0.2 – 0.3) of dodecane-thiol capped HgTe nanocrystals (dashed lines). We can see that gain coefficients of 100/cm are achieved across the entire telecommunication window (indicated with the standard pass bands O/E/S/C/L). These values correspond to material gains reported for competing technologies such as nanocrystalline silicon and epitaxial quantum dots. It would also provide sufficient gain to allow for amplification of long-range surface plasmon polaritons (SPPs) for e.g. optical interconnects or SPP based lasers.

The HgTe synthesis does not require complex colloidal hetero-structure fabrication or vacuum epitaxy, allowing large batch fabrication with high yield. Several procedures exist to incorporate HgTe in electrically active devices, either polymer-dispersed or embedded in inorganic matrices. This makes the use of HgTe colloidal NCs in integrated photonics viable. In addition, the ultralow threshold for stimulated emission could allow for electrically pumped nanocrystal laser. Indeed, as mentioned by Wood et al. gain lifetimes of ca. 100 ns are required at sub-1 exciton population to achieve DC electrical pumping, a feat achieved by the colloidal HgTe nanocrystals used in this work.

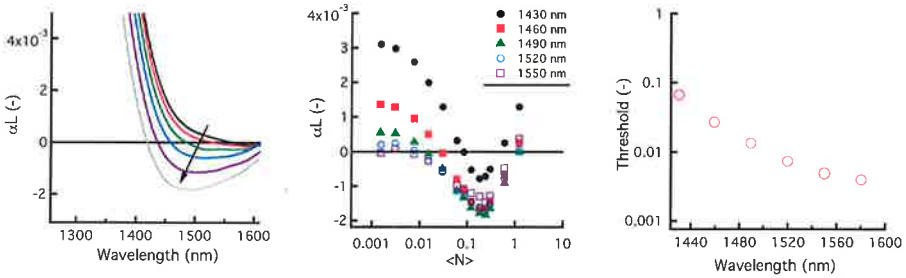

Figure 1: (left) Non-linear absorption spectra , taken 2.5 ns after photo-excitation for 1 size of HgTe nanocrystals. Stimulated emission corresponds to $\alpha L<0$. The maximum gain bandwidth extends from 1400 nm to beyond 1600 nm at sub-X fluence (<N>=0.3).(middle) Fluence dependence of stimulated emission as expressed by <N>, the average number of absorbed pump photons per nanocrystal at t=0 (right) Threshold for stimulated emission for different wavelengths throughout the gain band. The threshold is defined as the fluence <N> where αL becomes negative.

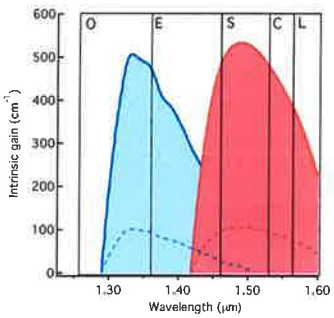

Figure 2: HgTe intrinsic material gain for two different samples emitting at 1220 nm (blue) and 1330 nm (red). The color-matching dashed lines indicate the volume-fraction corrected material gain. Note that the material gain provided by only 2 different sizes of HgTe covers the entire OESCL band with typical values over 500/cm.

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