Biexciton-mediated modulation response of colloidal quantum dots deposited on a silicon nitride waveguide at high laser excitation rate

M. Kolarczik[1], B. Herzog, C. Ulbrich, Y. Kaptan, U. Woggon and N. Owschimikow
Institut für Optik und Atomare Physik, Technische Universität Berlin, Berlin, Germany
A. Singh and X. Li
Physics Department, University of Texas at Austin, Austin, Texas, United States
Y. Zhu, D. v. Thourhout
Faculty of Engineering, Department of Information Technology, Ghent University, Belgium
P. Geiregat, Z. Hens
Faculty of Science, Department of Inorganic and Physical Chemistry, Ghent University, Belgium

[1]mirco.kolarczik@tu-berlin.de

Abstract: We develop a sensitive collinear pump-probe technique to resolve small amplitude and phase modulations of laser pulses in hybrid PbS/CdS-quantum-dot-SiN waveguides. It shows that at high excitation rate biexciton creation governs the optical interaction.
OCIS codes: (320.0320) Ultrafast optics; (340.7150) Ultrafast Spectroscopy

The integration of colloidal quantum dots (QDs) with silicon photonics is an exciting field of research, as it promises the realization of all-optical modulators and amplifiers. An important branch of modern optical telecommunications is the signal transmission and processing at high frequencies, which is currently largely based on solid-state semiconductor opto-electronics using on III-V semiconductors, for which exciton lifetimes are shorter than the transmission rates. To transfer these technologies to functionalized silicon waveguides will constitute a big step towards all-optical telecommunications. However, the long radiative lifetime of the ground state exciton typical for colloidal quantum dots represents a significant limit to a fast optical modulation response [1]. To quantify the influence of this long lifetime on the applicability of colloidal PbS/CdS QDs as an optical modulator in silicon photonics, we study the effect of a laser pulse train, at a high repetition rate of 75.4 MHz, propagating through a QD-coated (on top) silicon nitride [2]. Due to the quasi-constant offset created by the 1S exciton population, we expect the interaction to be mediated by nonlinear optical processes via the evanescent electric field of the laser pulses. The resulting small modulation ($\sim 10^{-3}$) is technologically important and challenging to measure.

Heterodyne-detected pump probe spectroscopy is a powerful tool to study physical processes in waveguides [3]. The heterodyning of the probe pulse with a reference oscillator allows one to work with pump and probe co-polarized and co-linear, a condition imposed by the waveguide, and still to extract pump-induced modifications of the probe pulse with high accuracy. A key feature of the method is that due to its interferometric nature it gives access to phase as well as amplitude changes. The noise floor is determined by laser power fluctuations and laboratory mechanical drifts and vibrations. In order to resolve very small signals otherwise obscured by these effects, in traditional pump-probe experiments a dual modulation of pump and probe is introduced, and detected by a lock-in detector set to the beat frequency. We transfer the sensitivity enhancement of this double-chop approach to the heterodyne pump-probe method, while still retaining the advantage of phase sensitivity, co-linearity, and co-polarization. Realizing that the probe modulation is readily replaced by the beating of probe and reference oscillator, we implement an additional sinusoidal fast amplitude modulation of the pump laser by an acousto-optical modulator. The resulting additional beat in the heterodyne signal in the 100 kHz range can be electronically well separated from the probe-reference beating. The setup is schematically displayed in Fig. 1 (a). Recording the standard heterodyne signal and its pump-modulation-induced sideband simultaneously with a digital dual lock-in detector provides an intrinsic referencing largely insensitive to laser fluctuations and laboratory drifts. This allows us to record a pump-induced differential transmission $\Delta S/S$ well below 10^{-3} at only few μW of probe laser power, which approaches the limit of quantum noise on the signal.

Fig. 1 (b) and (c) show the complex modulation response separated into differential changes of amplitude and phase of the probe laser pulse train. The probe laser wavelength is tuned to the 1S exciton resonance of the QD ensemble at 1200 nm (full width at half maximum ~ 20 nm), while the pump laser is tuned to a range of spectral windows on

the blue side of the peak, illustrated as inset in Fig. 1(c). We consistently observe a positive differential amplitude contribution, which corresponds to the pump pulse creating an additional population, which then amplifies the probe pulse. We assign this signal to biexcitons, which decay with a distribution of rates caused by dispersion in QD size and environment (inhomogeneous broadening). The extracted mean lifetime of around 150 ps, determined by a stretched-exponential fit agrees well with biexciton lifetimes in PbS/CdS QDs reported in the literature. Shifting the probe pulse to longer wavelengths still yields a positive change in amplitude. This indicates that the biexciton formation dominates over intraband absorption processes, which would cause a negative differential amplitude (absorption) of the probe pulse at short times. Biexciton formation is thus the dominating nonlinear optical process in colloidal PbS/CdS QDs, when working resonantly or quasi-resonantly with an ensemble of pre-excited dots.

(a)

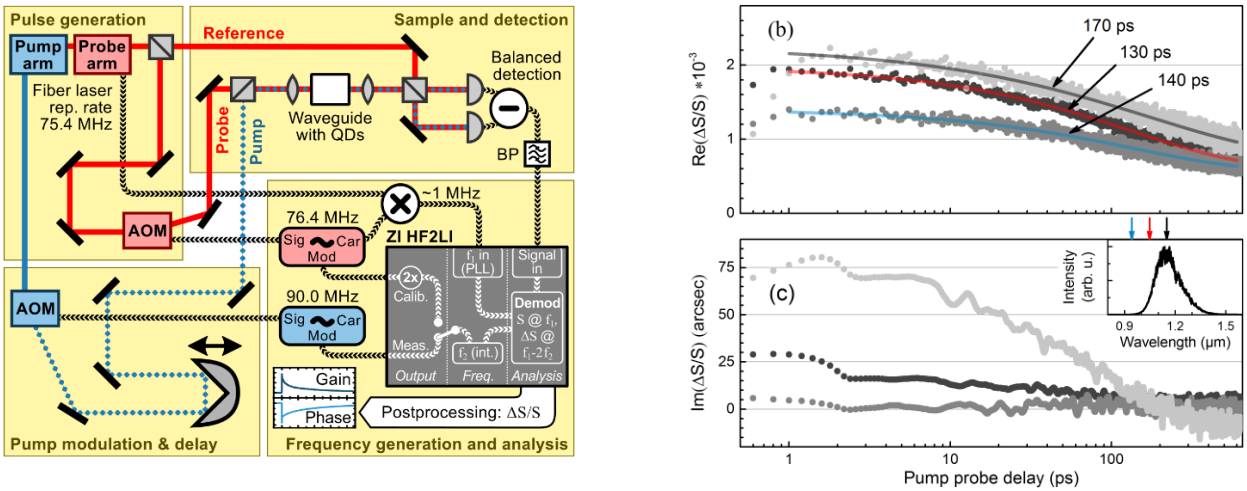

Fig. 1 (a) Experimental setup for the side-band heterodyne pump probe detection. (b) Real and (c) imaginary part of the differential transmission *ΔS/S*, representing the differential amplitude and differential phase, respectively. Experimental data is represented as scatter and the stretched exponential fit as solid lines with the extracted time constants listed above. The inset shows the photoluminescence of the QDs, on which the pump pulse spectral windows are marked as colored arrows.

References

[1] F. Masia, I. Moreels, Z. Hens, W. Langbein and P. Borri, "Four-wave-mixing imaging and carrier dynamics of PbS colloidal quantum dots", Phys. Rev. B **82**, 155302 (2010)

[2] A. Omari, W. Xie, P. Geiregat, D. V. Thourhout and Z. Hens, "Modeling the Optical Properties of Low-Cost Colloidal Quantum Dot Functionalized Strip SOI Waveguides", IEEE J. Sel. Topics Quantum Electron. **20**, 4400306 (2014)

[3] M. Kolarczik, N. Owschimikow, B. Herzog, F. Buchholz, Y. Kaptan, and U. Woggon, "Exciton dynamics probe the energy structure of a quantum dot-in-a-well system: The role of Coulomb attraction and dimensionality", Phys. Rev. B **91**, 235310 (2015)