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Functional nonlinear spectroscopy of nano-systems using phase modulated femtosecond light pulses

Dr. Khadga J. Karki
Lund University, Sweden

Abstract

The traditional methods of time resolved spectroscopy, e.g. the pump-probe and other forms of four-wave mixing experiments, allow us to investigate fundamental kinetic processes in atoms, molecules, molecular aggregates, and even macroscopic systems with femtosecond temporal resolution[1]. However, these measurements don't provide a clear answer to how the ultrafast processes contribute to the functioning of a photoactive device. Recent implementations of phase modulation schemes in optical excitation have addressed these limitations[2]. Phase modulated two-dimensional spectroscopy[3], in particular, provides a detailed picture of how the eigenstates in a system govern the ultrafast dynamics at femtosecond time scale and if such processes are relevant to the external signals such as photocurrent or photoluminescence. As the experiments are done by measuring the action of femtosecond pulses on the nonlinear photocurrent and photoluminescence response from the sample, measurements can be done directly on functioning devices. Moreover, these signals can be measured from systems with reduced dimensions, such as quantum dots, nanowires and even single molecules. Thus the new methods are expected to be used as the diagnostic tools to investigate the relationship between the tailored electronic structure and functionality in emerging nano-optoelectronic devices. I will give an overview of the recent developments and discuss future prospects.

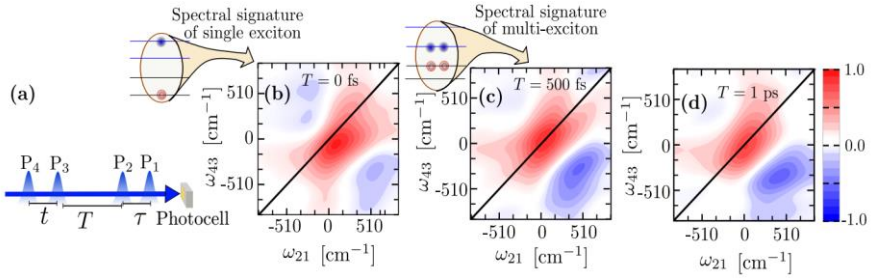

Figure 1: (a) Schematic of the photocurrent detected two-dimensional spectroscopy setup using phase modulation. (b,c,and d) Two-dimensional spectra of a PbS quantum dot photocell obtained by detecting the nonlinear photocurrent response. At zero time delay, $T = 0$ fs, the line shape is primarily absorptive (dominant positive feature along the diagonal), and at $T = 500$ fs, it becomes dispersive (positive and negative features separated by a nodal line). The lineshape remains dispersive at later time delays. The evolution of the dispersive line shape indicates that multiple exciton generation in the quantum dots contributes to the external photocurrent in the device (see ref. 2, Nat. Commun.).

[1] R. W. Boyd, Nonlinear Optics, Academic Press (San Diego, 2003)
[2] Karki et al., Nat. Commun., 5, 5869 (2014); Karki et al., Appl. Phys. Lett., 100, 153103 (2012); Karki et al., J. Opt., 18, 015504 (2015); Damtie et al., Phys. Rev. A, 96, 053830 (2017); Chen et al., ACS Photonics, 5, 1927 (2018).
[3] Tekavec et al., J. Chem. Phys. 127, 214307 (2007), Karki et al., arXiv:1804.04840v2.

ההרצאה תתקיים ביום רביעי, ה-15.5.19 בשעה 12:30
באודיטוריום המכון למצב מוצק, קומת כניסה

The lecture will take place on Wednesday, 15.5.19 at 12:30
at the Solid State Institute auditorium, entrance floor

Host: Assistant Professor Yoav Sagi