

Time-resolved photoelectron spectroscopy of lead sulphide quantum dots

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Understanding the details charge separation, charge transport and charge recombination at an atomic level is of high interest for emerging solar cell technologies such as those using thin films of semiconductor quantum dots. X-ray based techniques such as photoelectron spectroscopy (PES) are powerful tools for obtaining electronic structure information of materials at an atomic level. By combining photoelectron spectroscopy with a pulsed laser source, time-resolved pump-probe measurements can be carried out and electronic structure dynamics can be obtained: In time-resolved photoelectron spectroscopy (TRPES) a pump of visible or near infrared light is used to electronically excite the sample and X-ray radiation is used to emit electrons from the sample. The kinetic energy of the emitted electrons is measured and from this electronic structure and atomic level information of the sample can be gained (Figure 1). By varying the delay time between pump and probe pulses, the evolution of the electronic structure, which is induced by the pump pulse, can be followed over time.

This technique is of particular interest for solar energy conversion, as the pump pulse can mimic the sun light, which starts the photon-to-electron conversion process.

Timescales ranging from pico- to milliseconds combined with a large range of X-ray energies are available at synchrotron radiation facilities. These timescales match the timescales of charge transport, charge separation and charge recombination in quantum dot solar cells. In this presentation, I will show how such a methodology can be applied to PbS quantum dot films on n-type substrates.

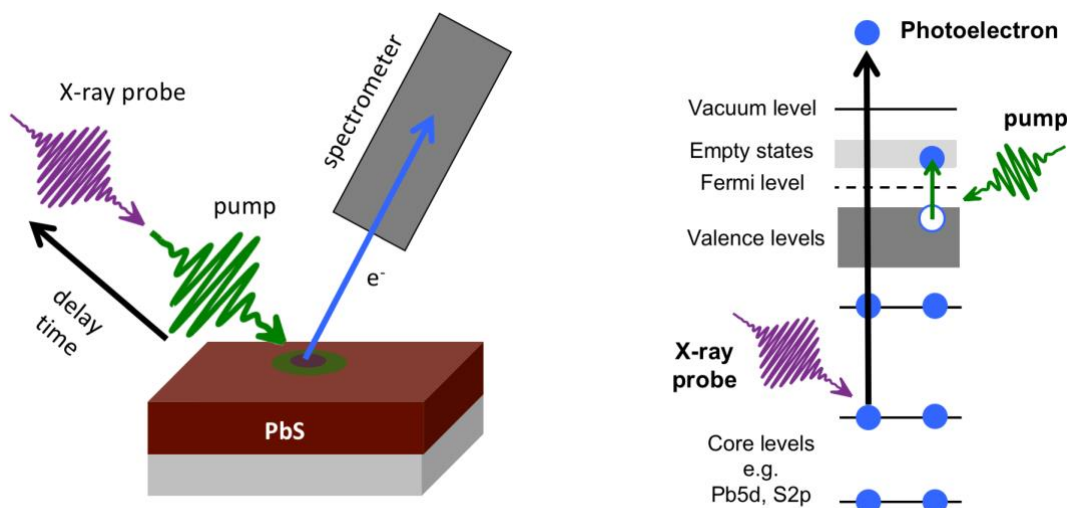


Figure 1: Concept of time-resolved photoelectron spectroscopy: Left: Schematic drawing of the experimental lay-out. Right: Electronic structure representation of TRPES. The change in core level positions induced by the pump laser is measured.

The measurements presented here were carried out at the LowDosePES endstation at Bessy II, where a highly efficient spectrometer can be used for photoelectron detection and a visible pump laser is available for pump-probe measurements [1]. The efficient spectrometer enables core level spectroscopy with short measurement times and makes time-resolved measurements feasible. The timescales of the dynamics, which can be studied with the set-up, range from the pulse length of the X-ray probe pulses (usually 70 ps) to the repetition period of the pump laser

(96 μ s in the experiments presented here). I will present results here from films PbS quantum dots with a lead iodide shell on Mg-doped ZnO (MZO) substrates. Electron injection from the PbS to the MZO leads to a shift of the Pb 5d core level to higher binding energies (Figure 2, left). By following the kinetics of this shift, we are able to follow the kinetics of the charge transport through the PbS film and the recombination of electrons back to the quantum dots (Figure 2, right). The presentation will show how these parameters depend on film thickness and laser intensity.

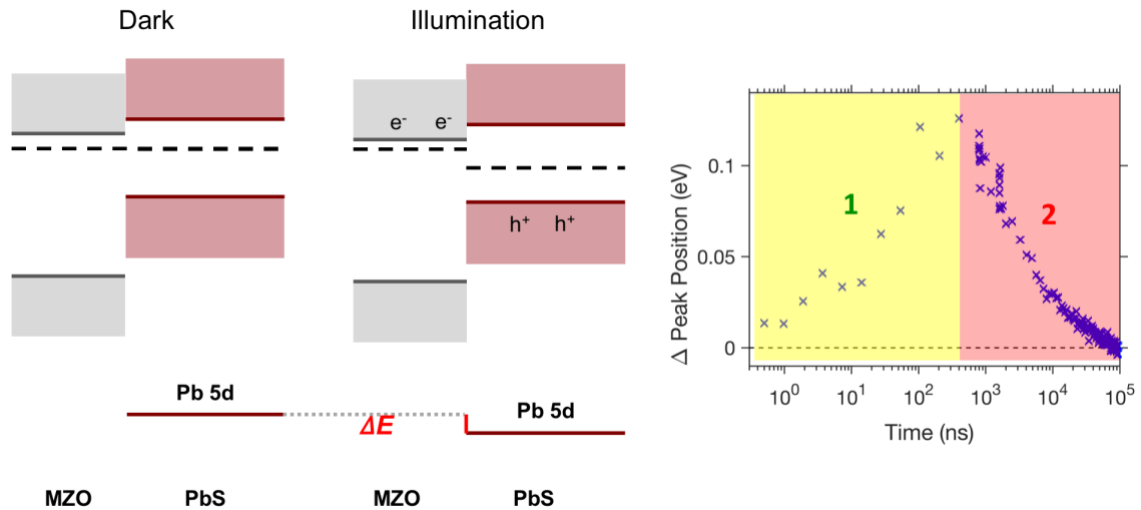


Figure 2: Left: Electronic structure of MZO/PbS interface showing how electron injection leads to a shift of the Pb 5d core level. Right: Shift in Pb 5d peak position of a PbS film on MZO as a function of pump-probe delay time. Region 1 highlighted in yellow shows the timescale of charge transport to the MZO substrate and region 2 highlighted in red shows the timescale of electron recombination.

References

- [1] E. Giangrisostomi, et al. *J. Electron Spectrosc. Relat. Phenom.*, **224**, 68–78 (2018).