Structure, Transport and Photoconductance of PbS Quantum Dot Monolayers Functionalized with a Copper Phthalocyanine Derivative

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Motivation

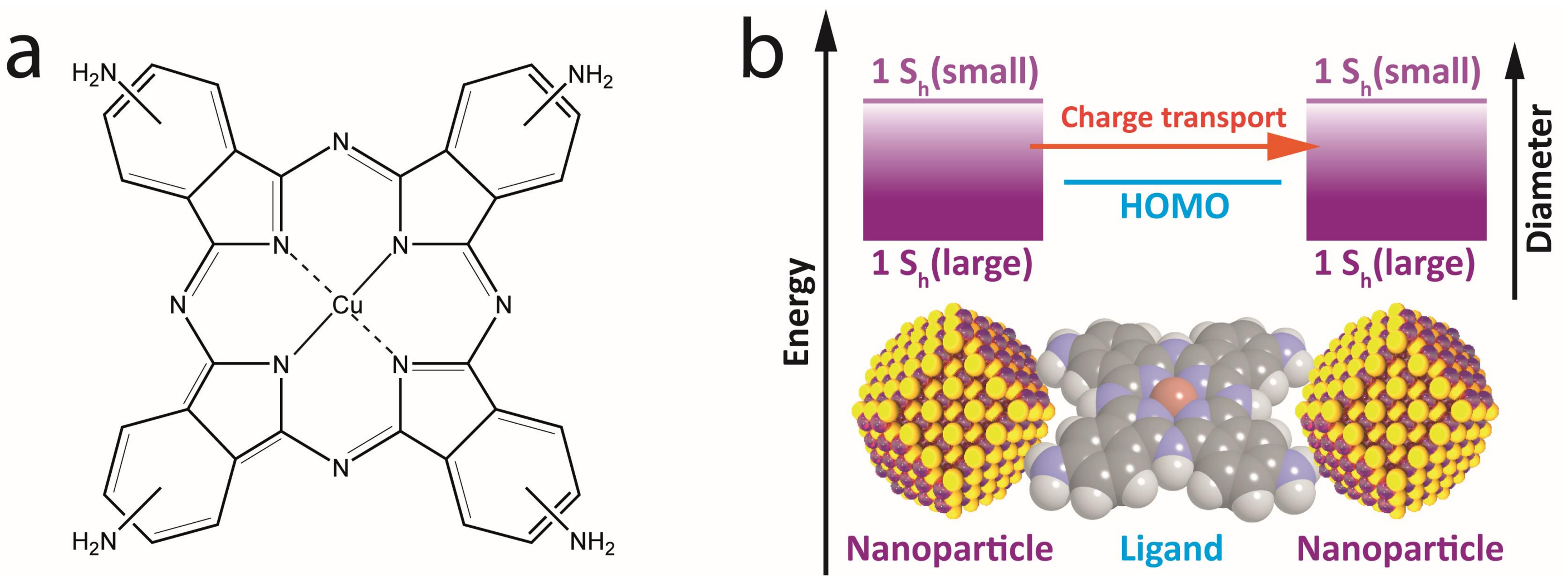
Combining semiconductor nanocrystals (NC) with organic semiconductor molecules into coupled organic-inorganic nanostructures (COIN) gives access to hybrid materials with entirely new properties.[1] We see great potential for a hybrid material comprising of PbS NCs and phthalocyanines (Pc) with many frequent interfaces for optoelectronic application.

The energies of the first excited hole state in PbS NCs and the highest occupied molecular orbital in Pcs are rather similar, which bears the prospect of resonant coupling for holes in this hybrid material.[2,3]

In this work, we fabricate films of PbS NCs functionalized with Cu 4,4',4'',4'''-tetraaminophthalocyanine (CuTAPc). We monitor ligand exchange at the NC surface, characterize the structure of the hybrid film, measure important electric transport parameters and assess the potential of this COIN for photodetection by building a light effect transistor.[4]

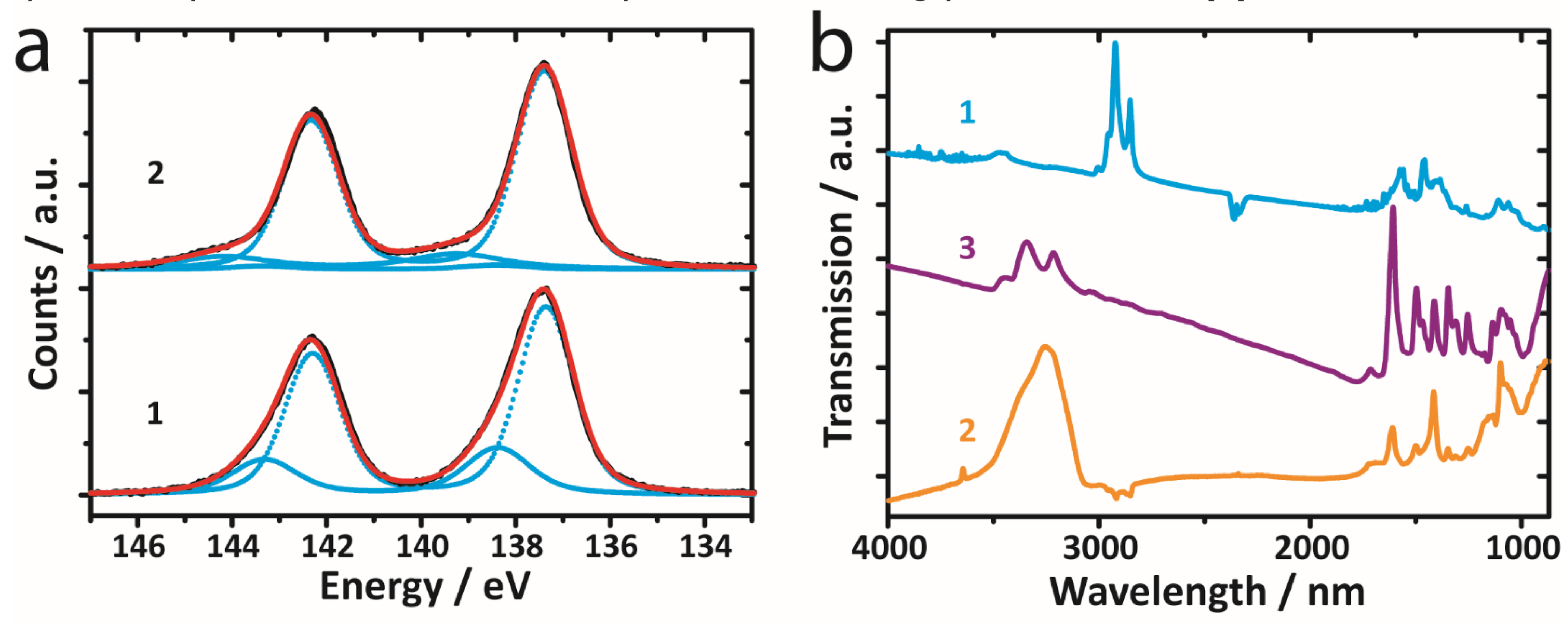
The Investigated System: PbS/CuTAPc

a) We use thin films of PbS NCs exchanged with Cu-Tetraaminophthalocyanine (CuTAPc) for COIN preparation. **b)** By carefully selecting the NC size we aim to improve the interaction between the NC's and the ligand's electronic states.



Ligand Exchange

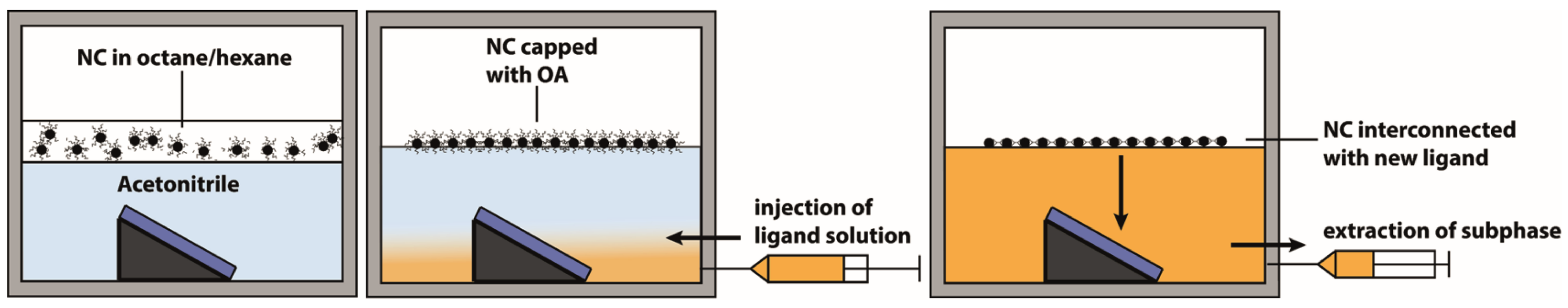
Ligand exchange is monitored via FT-IR and XPS*. **a)** XPS spectra of the Pb 4f PbS NC functionalized with Pb[Oleate]$_2$ **(1)** and after ligand exchange with CuTAPc **(2)**. **b)** FT-IR spectra of the same compounds including pure CuTAPc **(3)**.



*XPS data courtesy of Mahdi Samadi Khoshkhoo, University of Tübingen, Germany

Thin-Film Preparation Method

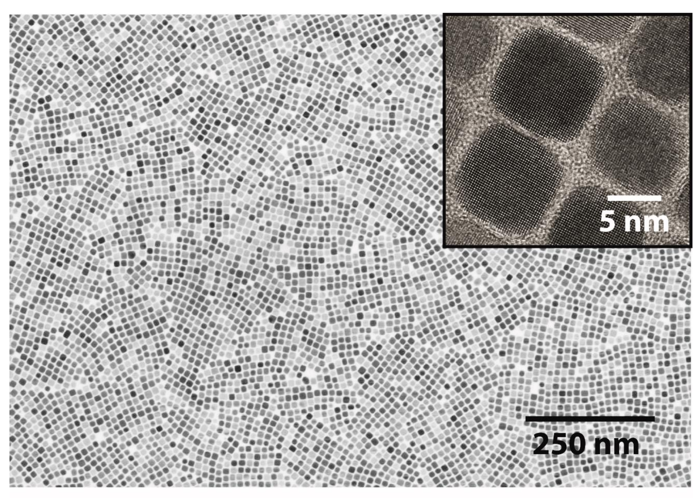
For film preparation and ligand exchange we make use of a Langmuir-type process of self-assembly at the liquid air interface.[5]



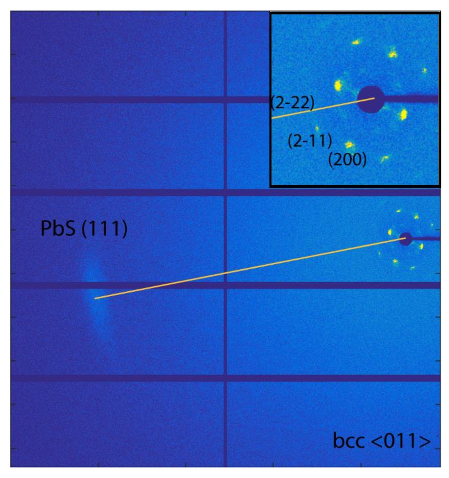
Morphology

The morphology of the functionalized films is investigated via TEM, GISAXS, SAXS and WAXS. The prepared films have a cubic superlattice (SL) and show significant angular correlation between the SL lattice planes and the atomic lattice in PbS.

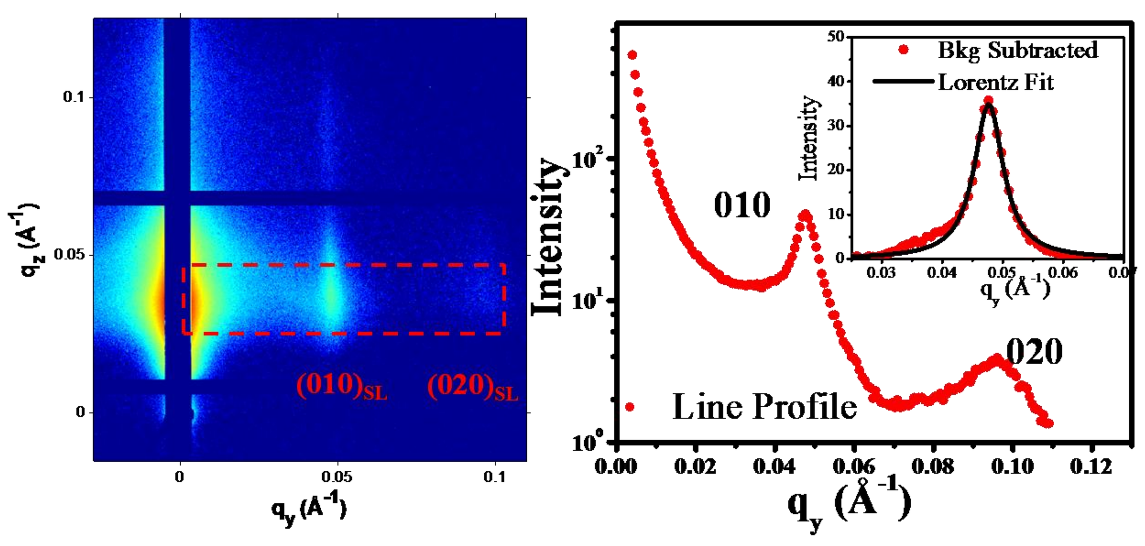
TEM/HR-TEM



Simultaneous SAXS/WAXS: shows oriented attachment.

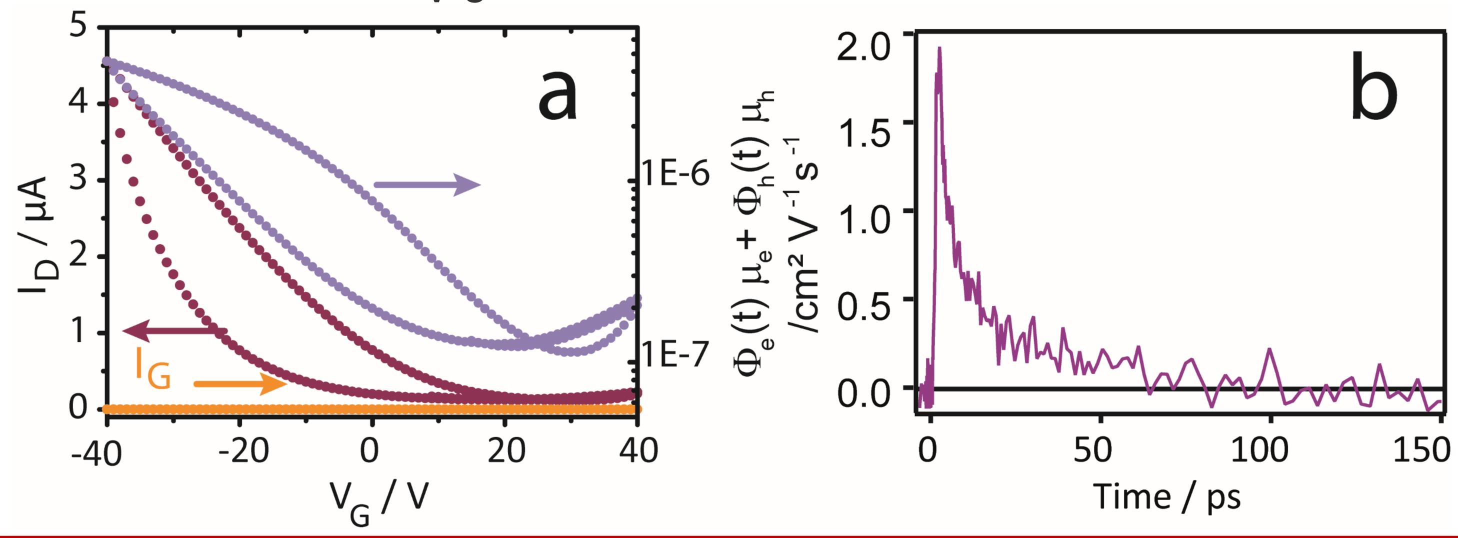


GISAXS



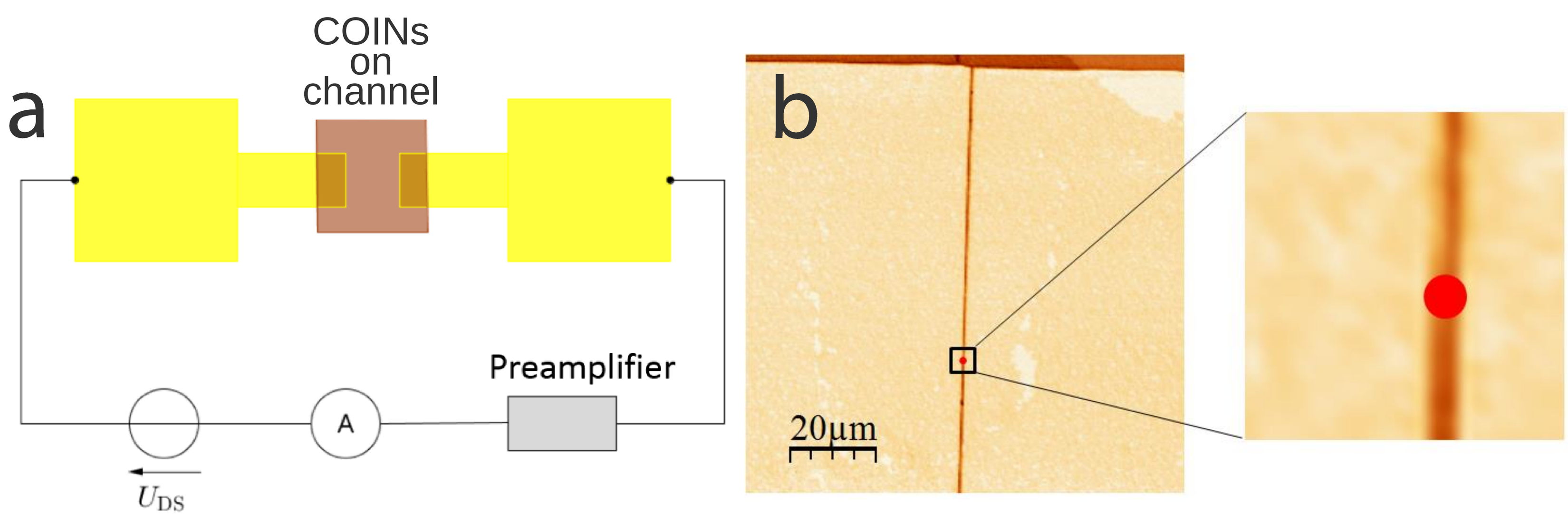
Charge Carrier Transport

a) FET measurements show hole transport, yield a mobility of $\mu = 10^{-3}\,\frac{cm^2}{V\cdot s}$ and a charge carrier concentration of the order of n = 1017 cm^{-3}. **b)** Terahertz spectroscopy yields a mobility of S(t) = 1 $\frac{cm^2}{V\cdot s}$.

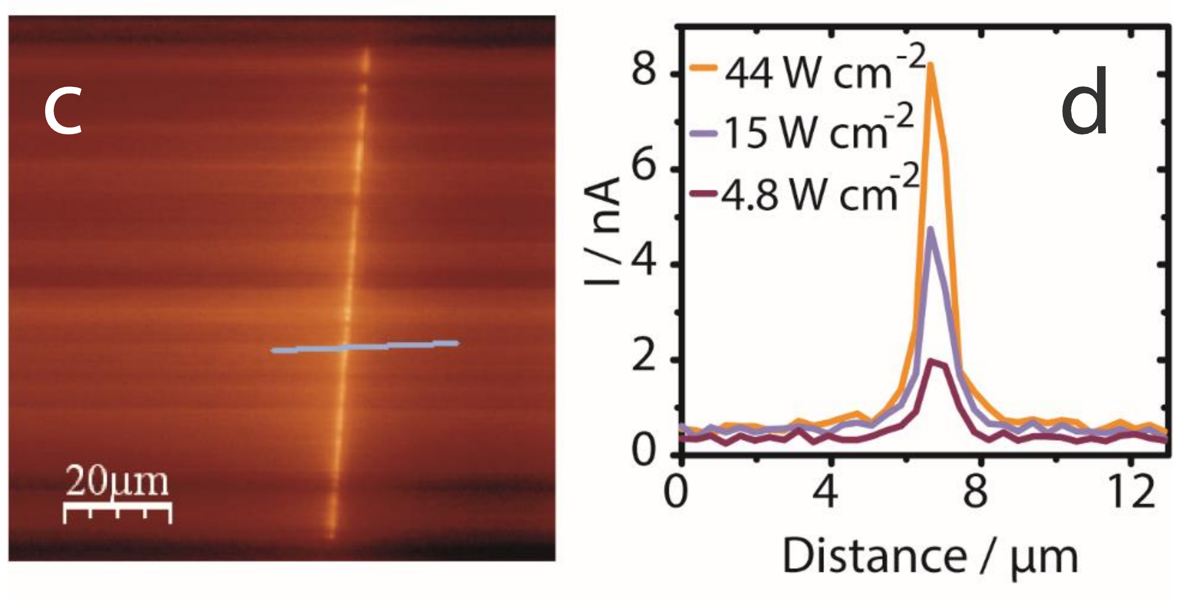


Light Effect Transistor

A light effect transistor is prepared by depositing a ligand exchanged COIN sample onto a test substrate with patterned gold contacts as depicted in **a)**. The channel is illuminated with a 633 nm He-Ne laser in a stage scanning confocal microscope as shown in **b)**.



The sample is raster scanned with the laser and the current across the channel is used as contrast information. As shown in **c)**, we measure a strong current modulation when the junction is illuminated. To verify this, we measure transport across the channel for different optical intensities and at constant bias of 3V **d)**.



Outlook and Conclusion

We prepared hybrid NC films of CuTAPc functionalized NCs. These monolayered films consist of mesocrystalline domains. THz spectroscopy reveals AC mobilities on the order of 1 $\frac{cm^2}{V\cdot s}$ over length scales similar to typical domain sizes. For larger length and time scales for which carrier scattering is relevant, the mobility is substantially reduced. We demonstrate the potential of COINs for light effect transistors and obtain an I_{on}/I_{off} ratio of ≤ 250 for an unoptimized device.

References

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