Sondervorträge

Four-wave mixing spectroscopy of exciton complexes in quantum dot molecules and 2D semiconductors

Dr. Daniel Wigger
Trinity College Dublin, The University of Dublin, Ireland

Investigating optical properties on the nanoscale: carrier density mapping and weak molecular absorption

Dr. Iris Niehues
CIC nanoGUNE BRTA, San Sebastian, Spain

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Aktuelle Fragen der Nanophysik

Freitag, 12.05.2023 um 15:15 Uhr
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Four-wave mixing spectroscopy of exciton complexes in quantum dot molecules and 2D semiconductors


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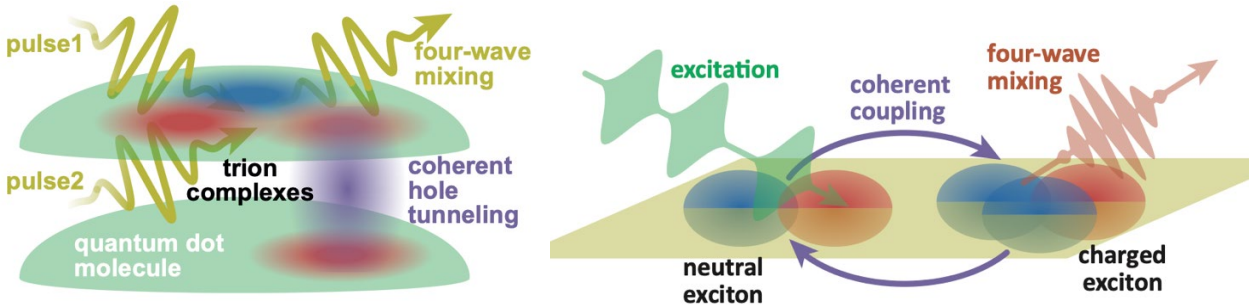
Trinity College Dublin, The University of Dublin, Ireland

Excitonic complexes in semiconductor nanostructures are widely considered as quantum resources due to their strong optical response. The processing technology for semiconductor quantum dots has been developed for a few decades and it is now possible to combine high-quality quantum dot molecules (QDMs) with photonic infrastructures and additional bias-tuning devices. This progress allows to reach a sophisticated control over the excitonic quantum states in such heterostructures. A more recent path explores the rich excitonic physics of layered 2D semiconductors, e.g., in the form of transition metal dichalcogenides (TMDCs). The encapsulation of TMDCs with hexagonal boron nitride improves th
e optical quality, while the combination with graphene allows to apply a gate biases.
While the linear optical responses of the two introduced systems are quite well understood, a deeper insight into their basic quantum dynamics, especially the interaction between different exciton complexed, e.g., neutral and charged excitons, requires nonlinear optical techniques like four-wave mixing spectroscopy.
In this talk I will shortly introduce the ultrafast four-wave mixing technique and present our recent studies on gate-tuneable QDMs [1] and TMDC monolayers [2]. In both cases we investigate in experiment-theory collaborations the coherent coupling between different exciton complexes and study how this interplay can be controlled by applying constant electric fields to the devices. In the QDM case the coherent coupling happens between two different trion configurations which interact via hole tunnelling between the two dots (left figure). In the TMDC case, the coupling happens between neutral and charged excitons where the density of the latter type can be enhanced with respect to the former one by the applied gate bias (right figure).



[1] D. Wigger et al., accepted in ACS Photonics (arXiv:2304.10148)
[2] A. Rodek et al., 2D Mater. **10**, 025027 (2023)

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Scattering-type scanning near-field optical microscopy (s-SNOM) and nanoscale IR point spectroscopy (nano-FTIR) allow for nanoscale optical mapping of manifold material properties. Both techniques are based on elastic light scattering at an atomic force microscope tip that is illuminated with monochromatic or broadband laser illumination. Acting as an optical antenna, the tip converts the illuminating field into a strongly concentrated near-field at the very tip apex. Interferometric recording of the tip-scattered field as a function of sample position yields near-field amplitude and phase s-SNOM images (employing monochromatic laser illumination), which encode information about the local dielectric function of the sample. while Fourier-transform spectroscopy of the tip-scattered field (employing IR broadband illumination) allows for nano-FTIR spectroscopy [1,2].

Intercalation of 2D layered semiconductors with molecules can drastically change the electric, optical, and magnetic properties of the host crystal. Recently, we found that MoS_2 bulk crystals become superconducting when intercalated with Tetraethylammonium (TEA) molecules [3]. Surprisingly, the superconducting state is not fully reached in few-nm-thick samples. To get a deeper understanding of the molecule distribution in the material we performed IR and THz s-SNOM and nano-FTIR spectroscopy to map the local carrier density of intercalated MoS_2. In the s-SNOM images we find a drop of the amplitude signals with increasing frequency and a change of the phase contrast, resembling a Drude-like response. Furthermore, the amplitude and phase images are not homogeneous, indicating a spatial variation of the local conductivity, i.e., the carrier concentration (Fig. 1). In addition, we use nano-FTIR to confirm the Drude-like response and to measure the molecular vibrations. By modeling the near-field spectra we can extract the local conductivity of the sample.

For dielectric samples such as biological materials or polymers, the near-field amplitude and phase signals of the scattered field reveal the local reflectivity and absorption, respectively. Importantly, absorption in s-SNOM imaging corresponds to a positive phase contrast relative to a non-absorbing reference sample. Interestingly, a negative phase contrast (NPC) can be observed when imaging a non-absorbing material on a highly reflecting substrate, which can hinder the recognition of materials exhibiting a weak infrared absorption (Fig. 2) [4]. We explore the origin of the NPC using representative test samples and demonstrate straightforward simple correction methods that remove the NPC and that allow for the identification of weak absorption contrasts.

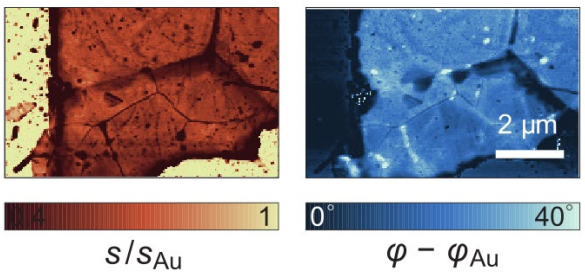

Figure 1: s-SNOM amplitude (left) and phase (right) image of the intercalated MoS_2 flake recorded at 1000 cm^{-1}.

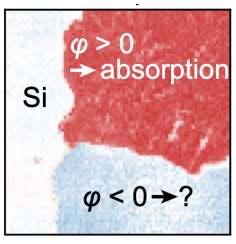

Figure 2: s-SNOM phase image of polymers on Si revealing positive (red) and negative (blue) phase values.

References

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[2] M. Schnell, et al., Nat Commun. 5 (2014), 349
[3] J. M. Pereira, et al. Adv. Funct. Mater. 208761 (2022)
[4] I. Niehues, et al. Opt. Express 31,7012-7022 (2023)