Experimentelle Physik 2

Seminar Festkörperphysik (CMP Seminar)

Aktuelle Probleme der Festkörperphysik für Studenten und Mitarbeiter
020236 Kolloquium/Seminar SS 24
Vorlesungszeit: 08.04.2024 – 19.07.2024

Thursday, 11.04.2024
12:00 - P1-O1-306

Long-lived exciton coherence and exciton-exciton interactions in bulk halide perovskite crystals

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Perovskite semiconductors are appealing for optoelectronic and photonic applications. The knowledge currently available about the energy structure of photoexcited electrons, holes and exciton complexes, their interaction, binding energy, and relaxation dynamics are far from being complete. Conventional time-integrated reflectivity or photoluminescence (PL) techniques alone often do not allow one to make unambiguous conclusions about the energy structure due to inhomogeneous broadening of optical transitions and complex dynamics of photoexcited carriers. Here, nonlinear optical techniques based on photon echoes or two-dimensional Fourier spectroscopy provide unique access to the energy structure of perovskite semiconductors.

We investigate the coherent dynamics of excitons in single bulk perovskite crystals. We observe exceptionally long exciton coherence time up to 80 ps at low temperature of 1.5K in mixed mixed-halide perovskite crystals due to the localization of excitons at the scale of tens to hundreds of nanometers [1]. The corresponding homogeneous exciton line width is 16 µeV, which is three orders of magnitude smaller than the inhomogeneous broadening (16 meV). From spectral and temperature dependences of the two- and three-pulse photon echo decay, we conclude that for low-energy excitons pure decoherence associated with elastic scattering on phonons is comparable with the exciton lifetime, while for excitons with higher energies, inelastic scattering to lower energy states via phonon emission dominates. Use of excitons with such narrow linewidth ensures an exceptional sensitivity for interaction effects on the µeV energy scale, e.g. probing the exciton-exciton interactions. In particular, polarization resolved transient signals provide rich information about the biexciton binding energy and spin dependent interactions in dense exciton ensembles [2].

[1] S. Grisard, A.V. Trifonov, I.A. Solovev, D.R. Yakovlev, O. Hordiichuk, M.V. Kovalenko, M. Bayer, and I.A. Akimov, *Nano Lett.* **23**, 7397 (2023).

[2] A.V. Trifonov, S. Grisard, A.N. Kosarev, I.A. Akimov, D.R. Yakovlev, J. Höcker, V. Dyakonov, and M. Bayer, *ACS Photonics* **9**, 621 (2022).