Optical properties of two-dimensional materials: equation-of-motion approach

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Abstract: Monolayers of transition metal dichalcogenide (TMD) semiconductors exhibit strong Coulomb interaction of their charge carriers giving rise to bound electron-hole states, known as excitons, with remarkable oscillator strength in optical spectra. Along with reciprocal-space valleys as a new optically addressable degree of freedom, this recommends TMD semiconductors as active materials in optoelectronic devices such as light-emitting diodes, solar cells, and lasers.

In my talk, I will first discuss the semiconductor Bloch equations (SBE), which represent an equation-of-motion approach to optical spectra in the time domain. In the linear regime, the SBE are equivalent to a Bethe-Salpeter equation. The equations allow to include many-body effects due to photoexcited electron-hole pairs such as band-structure renormalizations and phase-space filling. Moreover, nonequilibrium carrier kinetics after strong pulsed photoexcitation can be described by systematically extending the SBE.

In the second part, the SBE are applied to address the question how a finite density of excited carriers modifies the optical properties of TMD monolayers. Absorption spectra for excited carrier densities up to 10^13 cm^-2 reveal a redshift and mild bleaching of the excitonic ground state absorption, whereas higher excitonic lines are found to disappear successively due to Coulomb-induced band-gap shrinkage of more than 500 meV and binding-energy reduction. For investigations of the carrier kinetics in MoS2 I will present results for the carrier-carrier Coulomb scattering after pulsed optical excitation of the monolayer.