Unraveling the role of Coulomb interactions on the honeycomb lattice

Unusual series of excitonic resonances reveal an excitonic ground state in 2D transition-metal dichalcogenides.

Monolayers of transition metal dichalcogenides (TMDs) have recently attracted considerable interest, partially because of their unusual excitonic properties. Excitons, i.e. Coulomb bound electron-hole pairs, are the fundamental optical excitations in semiconductors. A number of recent linear und nonlinear optical experiments on a variety of TMDs observed excitons with unusually large binding energies and strong deviations from the Rydberg series known from conventional semiconductors. In J. Phys.: Condens. Matter 27 (2015) 345003 , we propose a theoretical explanation for the experimental observations that assigns the optical transitions to p-type excitations of a BCS-like exciton condensate.

Despite the close analogy between excitons and hydrogen atoms, there is one striking difference between them: Whereas atoms are real particles that exist in nature, excitons are quasi-particles in semiconductors that have to be created via absorption of photons. The optical resonances of hydrogen atoms occur at the energetic differences between the lowest 1s-state and the energetically higher p-states, while a typical GaAs-type semiconductor exhibits excitonic resonances at the energetic positions of the s-type excitons, usually beginning with the lowest 1s state.

In J. Phys.: Condens. Matter 27 (2015) 345003 , we explore the excitonic properties of TMDs treating the electronic quasi-particles as relativistic Dirac Fermions. A measure for the relative strength of the Coulomb interaction is the effective fine-structure constant α, and the critical value $\alpha=1$ marks the transition from a weakly to a strongly Coulomb interacing system. Unlike in quantum electrodynamics (QED) where $\alpha=1/137$ is a small parameter, the nominal value of α in TMDs ranges from 2-5/κ, where κ is the effective dielectric constant of the substrate/monolayer system. Depending on the properties of the substrate on which the TMDs are placed, the coupling can indeed exceed the critical value. As the fine structure constant relates the exciton Bohr radius a_B to the Compton wavelength λ_C via $a_B=\lambda_C/\alpha$, it is also a measure for the importance of relativistic effects. Therefore, we utulize the spinor eigenfunctions of the fully relativistic exciton problem. In the weakly interacting regime, we find a scenario like in usual semiconductors where the optical excitations correspond to s-type excitons with a total angular momentum of $|j|=1/2$, that are created by excitation of the noninteracting ground state. In contrast, in the strongly interacting regime, stable exciton resonances require $|j|>\alpha/2$, while states with $|j|<\alpha/2$ collapse under the strongly attractive Coulomb potential. In this regime, the Coulomb interaction is strong enough to form bound excitons *without* optical excitation, giving rise to a ground state with a BCS-like condensate of excitons. The dipole allowed excited states of such an excitonic insulator correspond to intra-excitonic transitions, just like the optical excitation of hydrogen atoms correspond to intra-atomic transitions. For the optically active states, we derive the optical selection rules $2|l|-1<\alpha<2|l|+1$.

Comparison of our theoretical predictions with recent experiments shows a remarkable agreement for the resonance positions, providing strong evidence for the realization of an excitonic ground state. In a way, our results imply that the observed excitonic series in TMDs are even more hydrogen-like than in a conventional semiconductor.

About the authors

The study was carried out in the Theoretical Semiconductor Physics group at the Philipps University Marburg. Stephan W. Koch is the group leader and full professor. His work focusses on the microscopic theory of optical properties of semiconductors and a selfsonsistent treatment of the relevant many-body effects. Tineke Stroucken is a post-doc in his group. She investigates novel 2D materials with a special focus on Coulombic effects on the ground state and optical response. The work has been supported by the Sonderforschungsbereich 1083 *Structure and Dynamics of Internal Interfaces*.