Exciton dynamics in atomically thin 2D materials

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Monolayers of semiconducting transition metal dichalcogenides (TMDs) build a new class of atomically thin two-dimensional materials. They exhibit a remarkably strong Coulomb interaction giving rise to the formation of tightly bound excitons. In addition to optically accessible bright excitonic states, there is also a variety of dark excitons that turn out to play a crucial role. To model these materials, we apply a microscopic approach combining the Wannier equation with TMD Bloch equations providing access to experimentally accessible excitonic absorption and photoluminescence spectra. In this talk, we review our recent work revealing the excitonic fingerprint of TMD materials:

(i) We provide a microscopic view on the time- and energy-resolved formation and thermalization of bright and dark excitons as well as their impact on the photoluminescence in different TMD materials. In particular, we demonstrate the crucial role of intervalley dark excitons.

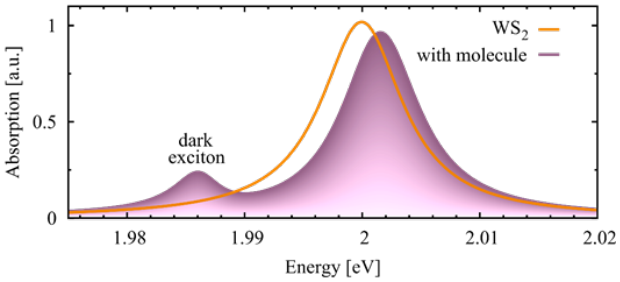

Fig. 1: Excitonic absorption spectrum of pristine and molecule-functionalized tungsten disulphide (WS_2). Efficient exciton-molecule coupling gives rise to a new peak that can be ascribed to dark exciton [4].

(ii) We present the excitonic absorption spectrum featuring a pronounced Rydberg-like series of excitonic transitions with binding energies in the range of 0.5 eV [1].

(iii) We investigate the microscopic origin of the homogeneous linewidth of excitonic transitions including radiative and phonon-assisted non-radiative relaxation channels as well as the appearance of exciton-phonon sidebands [2].

(iv) We show a significant disorder-induced coupling of bright and dark excitonic states offering a strategy to circumvent optical selection rules and make dark states visible in optical spectra of TMDs [3].

(v) We predict a novel sensor mechanism for molecules based on the molecule-induced activation of dark excitonic state resulting in the appearance of new well pronounced peaks in the absorption spectra of TMDs, cf. Fig. 1 [4].

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[2] M. Selig et al., Nature Commun. 7, 13279 (2016).
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[4] M. Feierabend, G. Berghäuser, A. Knorr, E. Malic, in print, Nature Commun. (2017).