2D materials for optoelectronic devices

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I. Introduction

There is currently interest in 2D transition metal dichalcogenide (TMDC) materials, MX_2 (M=Mo,W, X=S,Se,Te), for optoelectronic devices[1,2,3]. These materials, when thinned down to a single layer, are an example of atomically thin truly two dimensional direct gap semiconductors. The reduction of dimensionality is a reason for strongly enhanced electron - electron interactions, which result in optical properties at room temperature dominated by neutral and charged excitons with binding energies orders of magnitude larger than room temperature and those found in standard compound semiconductors, e.g., GaAs quantum wells.

The underlying honeycomb structure, as in graphene, results in the existence of two non-equivalent valleys, in which top of the valence and bottom of the conduction bands are located [4,5]. However, the two different atoms on the two sublattices result in opening of the gap, broken inversion symmetry and opposite topological moments associated with each valley. This results in coupling of each valley selectively with circularly polarized light. These valley dependent selection rules make MX_2 materials promising to use as polarized light absorbers / emitters and materials for valleytronics. This selective absorption / emission processes can be additionally controlled with carrier density. At low carrier density electrons can spontaneously occupy a single valley, in a valley polarized electron gas state [6,7]. Such material might lead to devices converting unpolarised to polarised light.

Another interesting feature of the band structures of TMDCs is the existence of a second minimum in conduction band, referred to as a Q point, which is associated with change of symmetry dictated contribution of orbitals to the conduction band between K and Gamma points in hexagonal Brillouin zone [5]. Around this point, the band nesting, i.e. parallel conduction and valence bands, result in very large joint optical density of states and strong enhancement of light-matter interaction by the so-called C-exciton. The enhancement of light-matter interaction is relevant for LEDs, lasers and light harvesting devices. The presence of C-excitons and reduced screening in 2D systems is also responsible for strong renormalization of low-energy excited excitonic spectrum, explaining strong deviation of series of optically active s-like excitonic states from the 2D like Rydberg series. Additionally, the non-trivial Berry's curvature in both conduction and valence bands affects the electron-hole pair states. This results in visible topological splitting of excited exciton 2p states, which can be detected by e.g. two-photon experiments.

Charged exciton (trion) is another interesting optically active complex in those systems. Due to enhanced electron-electron and electron - hole interaction, additional electron binding to the exciton is also very strong. Unusually, this binding energy of the trion is comparable with phonon energies in those systems, making them an unique platform to study exciton-trion-phonon interaction effects [8,9]. The strong binding energies of localised exciton complexes result also in room temperature upconversion of emitted light, a first step toward laser cooling at the nanoscale[9].

In this work we address challenges in theory of optical response of TMDC materials, focusing on excitons and using ab-initio based tight-binding electronic band structure and Bethe-Salpeter (B.-S.) equation for excitonic properties.

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II. Method

Throughout this work we use density-functional theory based tight-binding electronic structure model, discussed in details in Ref. [5]. By using symmetry arguments, we construct minimal graphene-like model, which allows us to capture all important features of ab-initio band structure. The material couples with light via excitons. The excitons, linear combination of electron-hole pair excitations, are solutions of B.-S. equation [10]:

$$\left[\Delta E(k)-\Delta-V_{\sin.}(k)\right]A_n(k)-\sum_{k'\neq k}^{BZ}(\delta k)^2 V(k,k')A_n(\vec{k}')=E_n A_n(k)$$

where $\Delta E(k)$ describes uncorrelated electron-hole pair energy, Δ – Kohn-Sham gap, $V_{sin.}$ – singular terms due to k=k' direct Coulomb interaction V(k,k'), $A_n(k)$ - exciton wavefunction coefficients in reciprocal space, E_n - binding energies and $(\delta k)^2$ is area of the Brillouin zone element coming from discretization of the k-space. The solution of Eq.1 gives ground and excited exciton energies E and corresponding wavefunctions A(k). Direct Coulomb interaction V(k,k') is computed using tight-binding wavefunctions with realistic approximation of localized orbitals using Slater – type basis and Keldysh - type screened interaction.

III. Results

Using MoS_2 as representative of TMDC's MX_2 material class, we solve Bethe-Salpeter equation for tight-binding dispersion and realistic interaction in each valley. This yields 1s exciton binding energies in the range of ~400 meV, consistent with experimentally reported values [2,6,8,11], and a spectrum of excited states differing from a simple 2D exciton, as shown on Fig. 1. Using simplified form of Coulomb interaction, we show how band nesting affects the low energy exciton spectrum. Then we focus on effect of tight-binding wavefunction form factors,

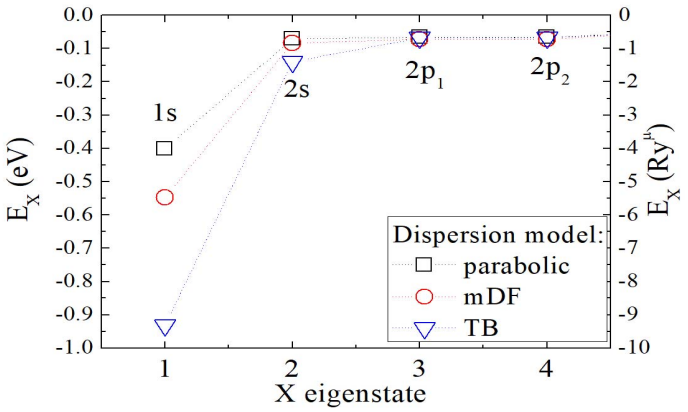

Figure 11: Effect of dispersion model on low energy spectrum of exciton.

resulting in topological moments which modify strongly electron – hole interaction V(k,k') and exciton spectrum. For a typical binding energy of 1s state of ~460 meV we obtain corresponding topological splitting of 2p dark exciton states of 13 meV.

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