Correlation effects in 2D Materials

Non-local manipulation of correlation effects in 2D Transition Metal Dichalgogenides

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In Short

- Non-invasive manipulation of electronic properties of 2D materials through *Coulomb engineering*.
- Compact unifying theoretical framework for metallic TMDCs.

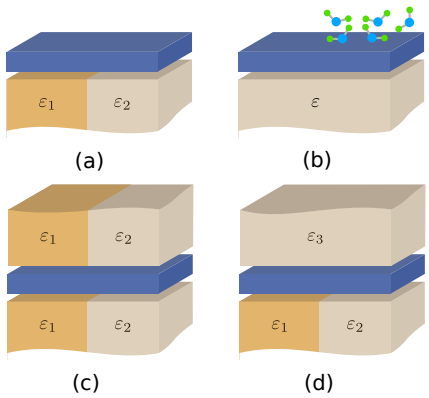

Figure 1: *Sketches of a monolayer (blue) in different laterally structured dielectric environments. (a), (c) and (d) show situations with structured substrates while in (b) adsorbed polarizable molecules are responsible for the heterogeneous dielectric environment. Reprinted from [1].*

Recent theoretical [1] and experimental works [2] have introduced the idea of a novel type of heterojunctions within two-dimensional (2D) materials based on the idea of manipulation of the Coulomb interaction through laterally structured substrates as depicted in Fig. 1. These so called *Coulomb-engineered* heterostructures show spatial property changes, like different band gaps, within an otherwise homogeneous monolayer.

For many applications the band alignment in heterojunctions is important. By means of ab-initio calculations we studied this alignment in Coulomb-engineered transition metal dichalcogenides (TMDCs) for different substrates. We see that we can have different alignments depending on the monolayer and substrate combination. How to manipulate these types of alignment by strain, defects or vacancies is still under investigation.

Not only the band alignment but also effects resulting from the interface of the substrates are of great interest. We study with G_0W_0 calculations monolayers partially covered with hBN. First calculations show unexpected and interesting results with large effects of edge states of the substrate, which need further studying. As these calculations are numerically very demanding we will apply material-realistic model approaches to understand the effects of spatially changing screening at the interface and away from it in homogeneous monolayers.

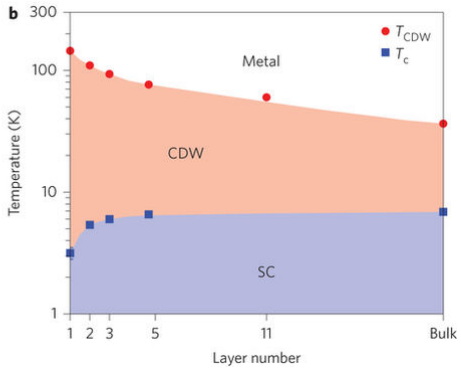

Figure 2: *Thickness-temperature phase diagram of $NbSe_2$ showing competing phases. Reprinted from [3].*

In metallic monolayer TMDCs, competing phases have been observed experimentally as for example shown by Xiaoxiang Xi et al. [3] in $NbSe_2$ (cf. Fig. 2). TMDCs constitute a prominent showplace for competing many-body instabilities such as superconductivity [4], charge-density waves [5] and magnetism [6] as well as Mott insulating phases [7]. In this context, we have shown that even though the observed phase diagrams are complex, the underlying mechanisms are captured by a compact unifying theoretical framework. We apply the constrained random-phase approximation (cRPA) [8] and constrained density-functional perturbation theory (cDFPT) [9] to the metallic monolayers *H-MX*$_2$ with $M \in \{\text{V, Nb, Ta}\}$ and $X \in \{\text{S, Se}\}$ and summarize the material specifics with a small number of representative Coulomb and electron-phonon interaction parameters.

In Fig. 3, cRPA estimation of the screening (dielectric function) and the screened local Coulomb interaction (minimal parameters through Kanamori parametrization) for all the metallic TMDCs are plotted. It exhibits a systematic behaviour of the strength of electronic correlations as one scans through different transition metals and chalcogens in TMDCs.

In Fig. 4, the application of cDFPT is visualized using the example of $NbSe_2$: Regular density functional perturbation theory (DFPT) [10] yields soft phonon modes, indicating structural instability, which

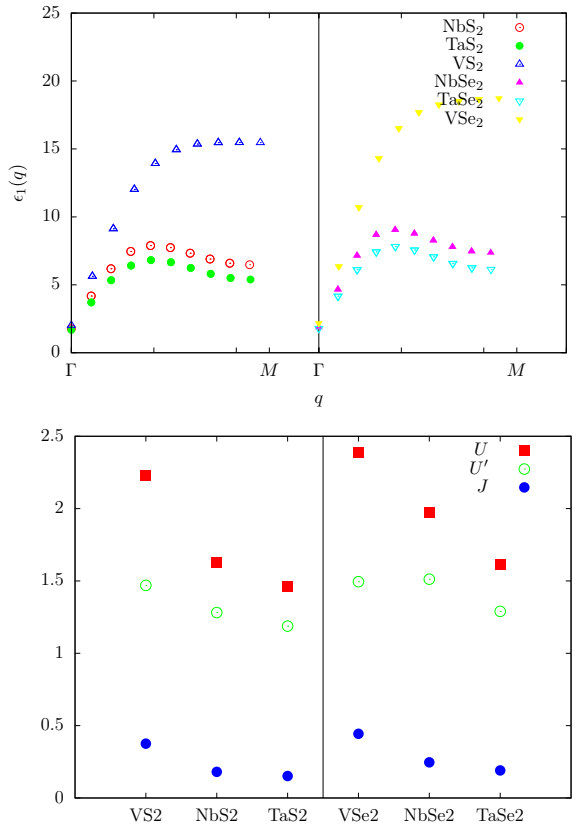

Figure 3: *(Top panel) q-dependent dielectric function $\epsilon_1(q)$ along the path Γ-M and (Bottom panel) Kanamori parametrization of the local Coulomb interaction for different TMDCs.*

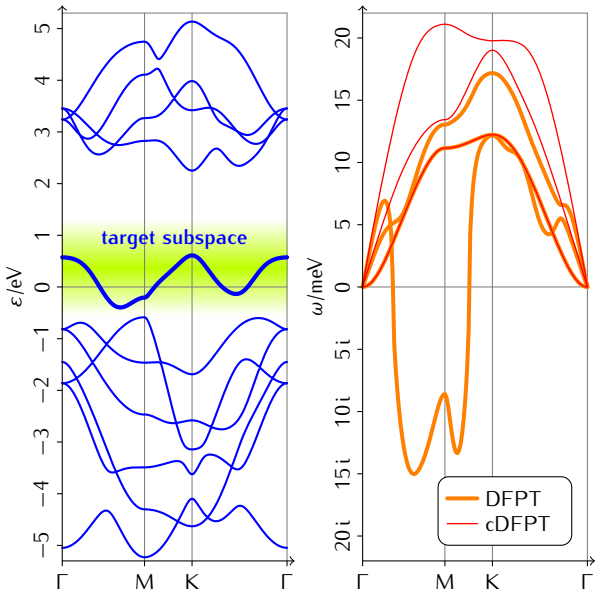

Figure 4: *Application of cDFPT to $NbSe_2$. Left: Electron dispersion. Right: Acoustic phonon dispersion according to (c)DFPT, including (excluding) the response of the target subspace.*

disappear upon exclusion of the metallic band from the electronic response to phononic lattice distortion.

Effective lattice models, constructed from available tight-binding Hamiltonians using cRPA and cDFPT parameters, will be further investigated with efficient many-body approaches like Quantum Monte Carlo or Green's function based perturbative schemes. These studies will provide a systematic theoretical description of the emergence of many-body instabilities in the metallic transition metal dichalcogenides.

We will also study the effect of screening on the Coulomb interaction as we go from bulk to monolayer thicknesses. This will provide a systematic evidence of whether or not free-standing monolayers are good examples of strongly correlated systems. Since the effect of screening is reduced in free-standing monolayers, we will also investigate the effect of the environment on the properties controlled by the Coulomb interaction like magnetism and superconductivity. Our findings could open up a whole new class of non-invasive applications and we want to guide experiments towards novel electronic as well as optoelectronic material functionalities based on manipulations of electronic interactions combined with spatial inhomogeneities.

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More Information

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