Excitons in van der Waals Heterostructures: The important role of Dielectric Screening

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The possibility of tailoring excitonic properties by vertically stacking two-dimensional layers in van der Waals heterostructures (vdWHs) represents a new paradigm in material science. Ab-initio calculations should in principle provide a powerful tool for guiding the design of vdWHs, but in their traditional form they are limited to commensurable structures with a few layers. Here, we overcome these limitations rephrasing the exciton many-body problem in terms of an effective 2D hydrogenic Hamiltonian (Mott-Wannier model) where the interaction between the electron and the hole is screened by the characteristic spatially non-local dielectric function of vdWHs. We calculate the dielectric properties using a multiscale approach where the dielectric functions of the individual layers (the dielectric building blocks) are computed ab-initio and coupled together via the long-range Coulomb interaction\(^1,2\). The dielectric functions of more than 50 different 2D materials are available in an open database together with the software for solving the coupled electrostatic equations\(^3\).

We demonstrate that while for monolayers and few-layers vdWHs the spatial non-locality of the dielectric function is well described by its linear approximation in reciprocal space\(^1,4,5\), for vdWHs where the exciton radius is comparable or smaller than their thickness, it is crucial to account for non-linearities. With a proper description of the dielectric screening, we are then able to calculate accurate exciton binding energies in agreement with experiments. As an illustration, we show in fig. 1, how the peculiar non-Rydberg exciton series in supported WS\(_2\), recently observed experimentally, is well reproduced by our calculations. Our method can even be applied to the design of more complex many-body excitations where the electron and the hole are located in distinct layers, namely inter-layer excitons (see fig. 2 (a)). Beside being affected by the dielectric environment, inter-layer excitons properties can be further tuned engineering the electron-hole separation. Figure 2 shows, for example, how the inter-layer exciton binding energy in a MoS\(_2\)-WSe\(_2\) based heterostructure can be varied over a broad range by simply intercalating a different number of h-BN layers.

In summary, our approach is accurate, yet highly efficient and has the merit of providing a seamless connection between the limit of isolated monolayer materials and the more complex case of van der Waals heterostructures.

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3. The dielectric building blocks and the QEH software can be downloaded from https://cmr.fysik.dtu.dk/vdwh/vdwh.html

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FIG. 1: Excitons in supported WS\(_2\). (a) Binding energies of the lowest five excitons in freestanding WS\(_2\) and WS\(_2\) on hBN (experimental data from 6). (b) The screened electron-hole interaction in WS\(_2\) monolayer adsorbed on hBN along with the radial probability distribution of the first five excitons.

FIG. 2: Inter-layer Excitons in MoS\(_2\)-WSe\(_2\) based heterostructures. (a) Schematic of the structure. (b) Binding energy of the lowest intra and inter-layer (green) excitons as function of the number of sandwiched hBN layers.