

# Charge-carrier localization in transition metal dichalcogenide nanostructures

## Interplay of structural, electronic, and optical properties in transition metal dichalcogenide nanostructures

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### In Short

- Semiconducting transition metal dichalcogenides are promising candidates for novel single-photon applications.
- Their opto-electronic properties are largely influenced by local strain and changes of the dielectric environment.
- We investigate the interplay of both effects by using a combined microscopic tight-binding and force field approach.

In this project we investigate the structural, electronic, and optical properties in semiconducting two-dimensional transition metal dichalcogenide (TMD) nanostructures with a focus on application as single-photon sources. To simulate the corresponding excited charge-carrier localization mechanism in such two-dimensional layers, local surface wrinkling driven by the bending rigidity of these materials as well as the resulting strain field are essential building blocks due to their pronounced influence on the band gap. Additionally, local changes of the dielectric environment modify the charge-carrier confinement due to the strong susceptibility of TMD monolayers to external manipulation. The interplay of surface wrinkling, strain-induced confinement, and local changes of the dielectric environment is systematically studied using a combined microscopic tight-binding and REAX force field approach [1].

In the following, we summarize results for MoS<sub>2</sub> nanobubbles that are formed when placing a monolayer of MoS<sub>2</sub> on another MoS<sub>2</sub> sheet used as a flat substrate. To cover surface wrinkling, strain-induced confinement, and local changes of the dielectric environment, we model a TMD layer with atomic resolution in a supercell approach. Based on valence force field simulations, new equilibrium positions of the individual atoms in the bended material are determined. For this part, we utilize a REAX potential, which is capable of accurately describing bond deformations under strain as well as continuous bond formation and breaking dynamics. The information about the displaced atoms is used in a tight-binding

electronic-state calculation for the supercell structure. Strain-induced band gap changes arising from the displaced atomic positions are included via a generalized Harrison rule. In our real space tight binding representation the energy gap varies spatially due to local changes in the strain. Additionally, when locally detaching a TMD layer from the substrate underneath a modified dielectric environment and electronic hybridization is expected. We include both effects into our model by changing individual tight-binding parameters based on GW calculations [1].

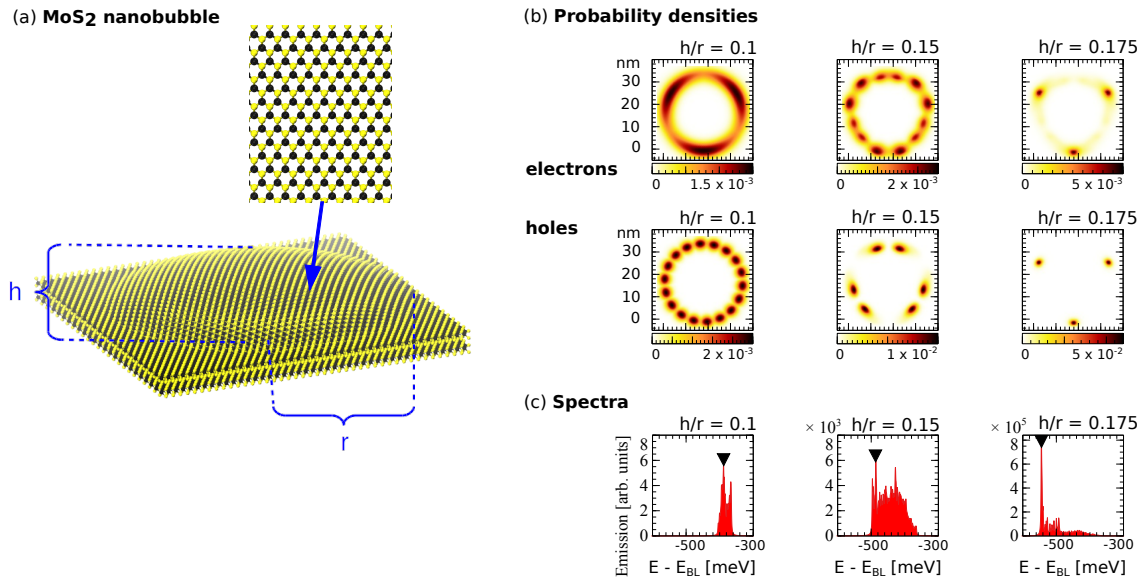
The degree of localization is controlled by the aspect ratio, as shown in Fig. 1(b). For  $h/r = 0.1$  the single-particle states are delocalized along the edge of the bubble, whereas for an increasing aspect ratio of  $h/r = 0.15$  distinct maxima are found. Due to the increased strain, these maxima carry the  $C_{3v}$  symmetry of the underlying crystal lattice. The trend of stronger localization with increasing aspect ratio continues for  $h/r = 0.175$ . In this case, the probability densities are localized at three distinct positions at the edge of the nanobubble. Figure 1(c) provides the emission spectra corresponding to the three different aspect ratios in Fig. 1(b), obtained from all confined states of the tight-binding model and Fermi's golden rule. For  $h/r = 0.1$ , the oscillator strength is very weak due to a small dipole interaction matrix element, which is the result of a much larger radial broadening of the probability density for the electrons than for the holes. More optical transitions are present for larger aspect ratios  $h/r$ . In the case of  $h/r = 0.175$  strong emission from the electron and hole states, shown in the right panel of Fig. 1(b), as well as a broad background emission from other states is obtained. This behavior can be explained by increasing strain, which leads to a deeper confinement, stronger localization of the single-particle states, as seen in Fig. 1(b), and a redshift of the optical spectra.

### WWW

<http://www.itp.uni-bremen.de/ag-jahnke/>

### More Information

- [1] Quantum-Dot-Like States in Molybdenum Disulfide Nanostructures Due to the Interplay of Local Surface Wrinkling, Strain, and Dielectric Confinement, *Nano Lett.* 19, 5 (2019)



**Figure 1:** (a) MoS<sub>2</sub> nanobubble with radius  $r$  and height  $h$ . (b) Top view of the probability densities for electrons and holes, which are responsible for the strongest optical transition marked in (c). Results are shown for a bubble radius of  $r = 18$  nm and various aspect ratios  $h/r$ . (c) Emission spectra calculated from Fermi's golden rule based on the tight-binding states for the aspect ratios in (b). Results are plotted with respect to the bilayer band gap.

### Project Partners

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