The goal of this project was to assess the impact of mechanical bending of two-dimensional transition metal dichalcogenides on their optoelectronic properties, using first-principles methods. These first-principles approximations for the electronic structure are built upon approximations within density functional theory and the quasiparticle GW, while methods for the optical properties are largely built upon many-body theory. GW-BSE is standard for optical absorption, but it is less practical for collective excitations as shown in model systems. Time-dependent density functional theory, however, has better promises for collective excitations in low-dimensional materials.

The work on two-dimensional (2D) materials has suggested the applicability of some meta-GGA functionals to band gaps and magnetic moments. Some meta-GGA functionals (TASK) have significantly more nonlocality in their exchange component than meta-GGA approximations designed for the ground state.

In my group we designed the mTASK approximation for low-dimensional materials where reduced screening plays a significant role in the electronic/optical response. We find that mTASK usually gives fundamental band gaps of the same quality as those of the HSE06 screened hybrid and improved magnetic moments in transition metal oxides as well as in CrI_3 nanoribbons.

mTASK can be a computationally more feasible alternative to HSE06 in band alignment and charge transfer calculations of large scale where conventional semilocal density functional approximations are not applicable.

The band gaps, optical absorption band gaps, and optical absorptions of nanoribbons are highly tunable. In our work we prefer bending instead of strain because the former can create effects that are beyond uniform strain (e.g., exciton funnel) In our recent work we take MoS_2 nanoribbons of different widths under bending and apply density functional theory and many-body perturbation GW and Bethe–Salpeter equation approaches. We find various response features that are more highly enhanced in nanoribbons than in bulk materials and even monolayers (nanoribbons are quasi-one-dimensional systems). Some characteristic response effects are attributed to the edge states that often produce in-gap states. The complex strain patterns on the bent nanoribbons control the varying features of band structures and band gaps that result in varying exciton formations and optical properties. The binding energy and the spin singlet–triplet split of the exciton forming the lowest absorption peak generally decrease with bending curvatures. The large tunability of optical properties of bent MoS_2 nanoribbons is promising and will find applications in tunable optoelectronic nanodevices.

We have identified bending as an effective control knob in phosphorene nanoribbons. We demonstrate the important role of the unoccupied in-gap state and conclude that this in-gap state in armchair nanoribbons introduced by bending can significantly affect the properties of low-energy excitons and add some useful opportunities for applications in optoelectronic devices.

The research on 2D materials has been extended to magnetic nanoribbons. In a recent work, we calculated band structures and magnetic moments of tensile-strained and bent zigzag CrI_3 nanoribbons with density functional theory. CrI_3 nanoribbons have a nearly equal preference for the out-of-plane and in-plane magnetic moment configurations, slightly favoring the in-plane one. We show that this enables four magnetization states in CrI_3 nanoribbons, including two out-of plane ones (up and down) and two in-plane ones (forward and backward along the nanoribbon periodical direction), increasing the operating controllability. Based on the one-dimensional Ising spin chain model, the spin correlation length of the

narrow CrI_3 nanoribbon was estimated to be about 10 A at its estimated Curie temperature of 27 K, which is lower than the measured 45 K of the monolayer CrI_3.

We also calculated optical absorption with many-body perturbation GW-BSE (Bethe-Salpeter equation) and investigated magneto-optical properties, including magnetic dichroism, Faraday and magneto-optical Kerr effects. Tensile strains and bending manifestly modulate the absorption spectra and magneto-optical properties of CrI_3 nanoribbons within a technologically important photon energy-range of ~1.0-2.0 eV, suggesting a potential application in tunable magnetic optoelectronics.

The low-energy dark excitons are mainly from transitions between electrons and holes with unlike spins and are non-Frenkel-like, while the bright excitons have mixed spin configurations. Using the GW-BSE method the intrinsic lifetime of excitons can be over one nanosecond, suitable for quantum information processes.

In a previous work (H. Tang, B. Neupane, S. Neupane, S. Ruan, N.K. Nepal, A. Ruzsinszky, Scientific Reports, 12, 3008 2022) we demonstrated that the edge bands evolved with bending can tune the optical properties for various widths of transition metal dichalcogenide (TMD) nanoribbons. Defects are commonly present in 2D TMD materials and can dramatically change the material properties. In this following work, we investigated the interaction between the edge and the defect states in tungsten disulfide (WS_2) nanoribbons with W - or S-atom vacancy defects under different bending conditions, using density functional theory. We combined in-gap defect states and in-gaps states from bending to provide a controlled phase transition in 2D materials.
These results broaden the device design strategies for WS_2 homojunctions and have potential applications in phase-change electrical devices.

We also confirmed that the appropriate choice of the defect sites can induce magnetic properties on the WS_2 nanoribbons. To gain understanding about the limits of density functional approximations, we compared results on band gaps and energies of defect states with quasiparticle GW. We reveal interesting semiconductor-metal phase transitions, suggesting potential applications in nano-electronics or molecular electronics. We also calculated the optical absorption of the bent and defective nanoribbons with the many-body GW-BSE (Bethe-Salpeter equation) approach, revealing a tunable optical spectrum and diverse exciton states in the defective WS_2 nanoribbons.

A WSe_2 monolayer shows many interesting properties due to its spin–orbit coupling induced spin splitting in bands around the Fermi level and the spin–valley configuration. With *ab initio* techniques, we found that the WSe_2 nanoribbons can exhibit an enhanced spin-orbit coupling (SOC) effect and a spatially varying spin-polarization in bands around the Fermi level under bending conditions. The spin-polarization can show an anisotropy (or asymmetry) in these nearly degenerate bands, leading to a controllable magnetism *via* bending and electron/hole doping of the nanoribbons, suggesting a potential application in compact and controllable magnetic nanodevices and spintronics. The optical absorption spectrum of the nanoribbon presents a large tunability with bending within the near infrared region of about 0.4 to 1.5 eV, showing an enhanced absorption under a large bending condition. The exciton states generally show mixed or various spin configurations in the electron and hole pairs that are controlled by bending, and are potentially useful for applications in spin-based quantum information processes.

Some of the 2D monolayers were predicted to have topological nontrivial edge states, due to band inversion around the Gamma point. Those topological nontrivial edge states are robust and can be harnessed for many novel applications. Some of these materials have been experimentally realized and confirmed ($MoTe_2$, WTe_2). Nanoribbon forms of those materials are often more attractive for industrial applications. Nanoribbons are less explored. Due to physical edges, edge interactions, and quantum confinement, the

edge states of nanoribbons are far more complex than the corresponding monolayers. Our recent work aims to understand the evolution of the edge states with the width, and orientation of nanoribbons, and considers the modification of the ribbon edges. Our ultimate goal is to establish a correlation between the optical and catalytical properties of those nanoribbons.

Model kernels within linear response time-dependent density functional theory (TDDFT) indicate a strong potential in the low-density regime with accuracy to describe quantum phenomena at the macroscopic level. The accuracy of TDDFT, however, largely depends on the approximations for the exchange-correlation (xc) kernels. Away from the long-wavelength (or $q = 0$ short wave-vector) and zero-frequency ($\omega = 0$) limit, the correlation contribution to the kernel becomes more relevant and dominant over exchange. The test charge-test charge dielectric function in principle can encompass xc effects relevant to describe low-density physics. Furthermore, besides collective plasmon excitations, the dielectric function can reveal collective electron-hole excitations, often dubbed "ghost excitons." Beside collective excitons, the physics in the low-density regime is rich, as exemplified by a static charge-density wave that was recently found for $r_s > 69$, and was shown to be associated with softening of the plasmon mode. Within this project we performed a thorough analysis with xc model kernels for excitations of various nature. The uniform electron gas as a useful model of real metallic systems is used as a platform of our analysis. We highlight the relevance of exact constraints as we display and explain screening and excitations in the low-density region.

We also performed an analysis regarding the nature of excitations in the plasmon and particle-hole continuum regions using the spectral function. We demonstrate that excitations within the particle-hole continuum maintain collective nature.

These results project the applicability of model xc kernels for real materials. Currently we are finishing a manuscript about collective excitations in $TiSe_2$. We find that, with the Drude term included, xc kernels are reasonably good predictors of the charge-density wave phase transition at low temperature. These xc kernels have high potential in further applications of charge and spin waves.

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