Electronic, optical, and magnetic properties of single-layer transition metal dichalcogenides in the presence of defects or impurities

Michael Leuenberger

Department of Physics, University of Central Florida, Orlando, FL

A detailed first-principle study has been performed to evaluate the electronic and optical properties of single-layer (SL) transition metal dichalcogenides (TMDs) (MX_2; M= transition metal such as Mo, W and X= S, Se, Te), in the presence of vacancy defects (VDs). Defects usually play an important role in tailoring electronic, optical, and magnetic properties of semiconductors. We consider three types of VDs in SL TMDs (i) X-vacancy, (ii) X_2-vacancy, and (iii) M-vacancy. We show that VDs lead to localized defect states (LDS) in the band structure, which in turn give rise to sharp transitions in in-plane and out-of-plane optical susceptibilities, $\chi_{\|}$ and $\chi_{\perp}$. The effects of spin orbit coupling (SOC) are also considered. We find that SOC splitting in LDS is directly related to the atomic number of the transition metal atoms. Apart from electronic and optical properties we also find magnetic signatures (local magnetic moment of $\sim \mu_B$) in $MoSe_2$ in the presence of Mo vacancy. We show that a simple qualitative tight binding model (TBM), involving only the hopping between atoms surrounding the vacancy with an on-site SOC term, is sufficient to capture the essential features of LDS. In order to provide a clear description of the optical absorption spectra, we use group theory to derive the optical selection rules between LDS for both $\mathrm{Im}\chi_{\|}$ and $\mathrm{Im}\chi_{\perp}$.

Room-temperature superparamagnetism due to a large magnetic anisotropy energy (MAE) of a single atom magnet has always been a prerequisite for nanoscale magnetic devices. Realization of two dimensional (2D) materials such as single-layer (SL) MoS_2, has provided new platforms for exploring magnetic effects, which is important for both fundamental research and for industrial applications. We use density functional theory (DFT) to show that the antisite defect (Mo_S) in SL MoS_2 is magnetic in nature with a magnetic moment of $\mu \sim 2\mu_B$ and, remarkably, exhibits an exceptionally large atomic scale MAE=$\varepsilon_{\|}$-$\varepsilon_{\perp}$ of $\sim$ 500 meV. Our calculations reveal that this giant anisotropy is the joint effect of strong crystal field and significant spin-orbit coupling (SOC). In addition, the magnetic moment μ can be tuned between $1\mu_B$ and $3\mu_B$ by varying the Fermi energy ε_F, which can be achieved either by changing the gate voltage or by chemical doping. Our systematic investigations deepen our understanding of spin-related phenomena in SL MoS_2 and could provide a route to nanoscale spintronic devices.

Ab-initio calculations for the electronic and optical properties of single-layer (SL) tungsten disulfide (SL WS_2) in the presence of substitutional Erbium impurities (Er_W) are presented, where the W atom is replaced by an Er atom. Although Er is much larger than W, we show that Er:SL WS_2 is stable using DFT. In order to understand impurity related optical transitions, odd states, which are usually neglected for pristine cases, need to be considered in addition to even states. We use group theory to derive strict selection rules for the optical transitions, which are in excellent agreement with the absorption spectrum calculated using the Kubo-Greenwood formula using the Kohn-Sham orbitals. Impurities usually play an important role in tailoring electronic and optical properties of semiconductors. We show that neutral Er_W impurities lead to localized defect states (LDS) in the band structure due to the f-orbital states of Er, which in turn give rise to sharp optical transitions in in-plane and out-of-plane components of the susceptibility tensor $\mathrm{Im}\chi_{\|}$ and $\mathrm{Im}\chi_{\perp}$. We identify the optical transitions at 5.3 μm, 1.5 μm, 1.2 μm, 920 nm, 780 nm, 660 nm, and 550 nm to originate from Er_W impurity states. Our results for the optical spectra are in good agreement with experimental data.