A theoretical study of atomic wires and atomic junctions created on a Molybdenum disulfide surface

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Introduction

The powerful drive for enhanced computational power is swiftly driving the electronics industry towards the atomic realm, where clusters of atoms, molecules and even single atoms act as the primary units for fabricating wires, switches, transistors and storage devices. Distinct from classical electronic devices, molecular and atomic electronics are governed by quantum mechanical principles, and thus understanding the transmission properties of these devices at the nano scale poses a scientific challenge. Experimentally, the scanning tunneling microscopy (STM) offers the possibility of manipulating and visualizing single atoms and molecules with atomic precision, and has facilitated the controlled fabrication of atomic structures.[1] [2] One such structure that has attracted much attention are atomic wires fabricated on the surface of a solid substrate; these wires pose a pathway to access molecules and atomic structures that can ultimately act as quantum logic gates. Moreover, recent experiments have shown that small metal islands deposited on surfaces can be manipulated by the STM; [3] these metal clusters can then act as nano-electrodes, and can be used to contact exactly one atomic wire thus offering the possibility to characterize the electrical properties of the wire through them.[4]

Dangling bonds created by removing atoms from a surface has been shown to possess promising conductance properties. These surface structures might replace molecular or oligomer wires, which are difficult to align reproducibly on the surface and difficult to contact externally. Molybdenum disulfide (MoS_2) presents an attractive candidate surface for the manufacturing of these atomic dangling bond wires, and as a substrate for a number of other atomic structures with interesting electronic properties. Controlled extraction of single sulfur atoms by a STM tip has been experimentally demonstrated.[2] In this project, we investigate from first principles the mechanisms governing the conductance properties of an atomic wire created by sulfur vacancies on a MoS_2 substrate. Furthermore, we investigate the physics behind a multilevel atomic junction formed by reinsertion of S atoms into an existing S vacancy wire.

Approach

State of the art density functional theory[5] was used to obtain electronic band structures and accurate density of states for the periodic MoS_2 structure and all the modified surfaces. The conductance properties of the systems were studied using the elastic-scattering quantum chemistry (ESQC) method,[6] which describes using quantum chemistry techniques the scattering of electron waves by a defect embedded in a periodic system. In addition, extended Hückel molecular orbital descriptions were employed to gain a more qualitative understanding of the electronic picture of the system. The familiar language of molecular orbital theory and band structure theory provide a convenient framework to discuss the results.

Results

Sulfur Vacancy Atomic Wire

It has been theoretically demonstrated that a finite atomic line of S vacancies created on a planar MoS_2 substrate can function as a pseudo-ballistic wire for electron transport.[4] This wire can be formed by extracting S atoms from the MoS_2 surface.

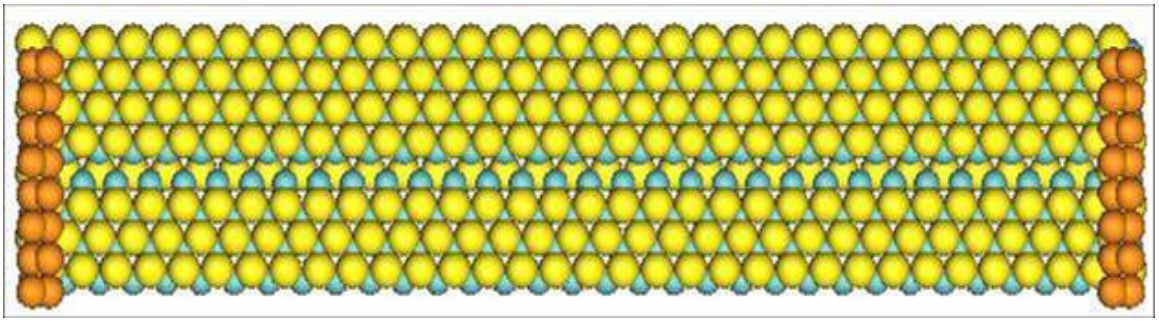

Figure 1. Model of a 10 nm atomic wire between 2 Au nano electrodes, created by extracting S atoms from a MoS_2 substrate

Removing surface S atoms introduces electronic states within the surface MoS_2 electronic band gap. Each S vacancy introduces a symmetric molecular orbital of mainly Mo d_z^2 character near the top of the valence band and a degenerate pair of molecular orbitals of mainly Mo d_{xz} and d_{yz} character near the center of the band gap. Mixing of the degenerate orbitals on neighboring vacancies causes the gradual formation of two overlapping energy bands near the Fermi level of the electrodes which create pseudo-ballistic electron transport channels.

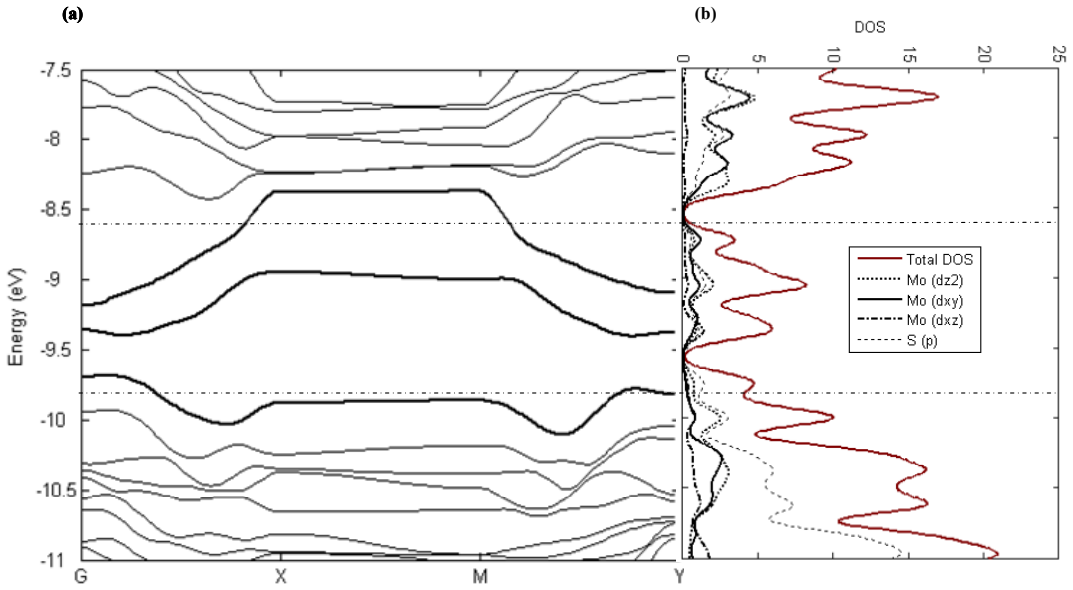

Figure 2: a) Band Structure for an infinite wire of S vacancies. Three bands (bold) characteristic of the wire are located within the original MoS_2 energy gap (dashed line). The upper two bands are responsible for the conductance. b) Density of States for the wire decomposed on atomic orbitals of different symmetry.

The conductance of the wire is independent of its length (Figure 3), but the overall channel transparency is not reaching unity, since electron waves are scattered between the Au nano electrodes and the wire of S vacancies. The conductance of an Au electrode – atomic wire – Au electrode junction can be enhanced by optimizing the Au electrode-wire distance and by increasing the lateral Au electrode-wire overlap by sliding the electrode over the wire.

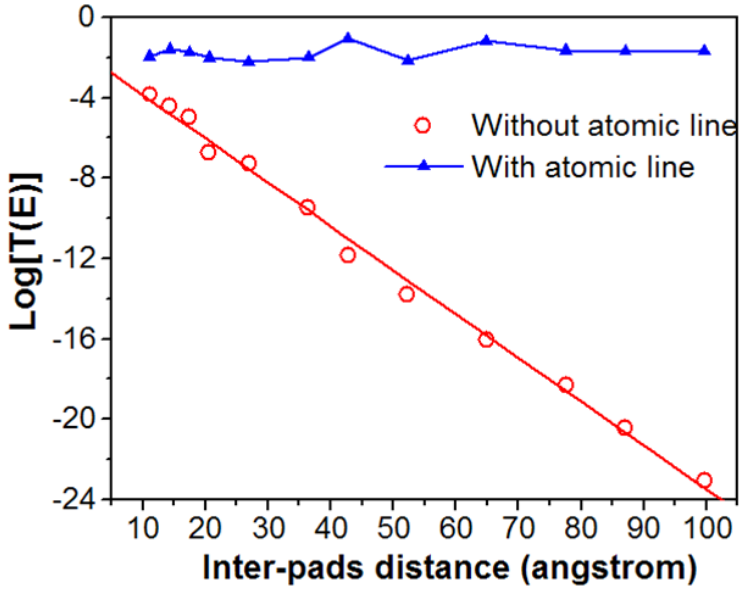

Figure 3: Variation of the transmission coefficient at the Fermi level as a function of the distance between the two Au nano-pads for a surface MoS_2 junction (o) and for an atomic wire of S vacancies fabricated on the MoS_2 surface (▲).

Atomic Junction

Taking the concept of the atomic wire one step further, we studied the electronic properties of atomic junctions of various lengths that can be created by reintroducing S atoms in the wire (Figure 5). The introduced S atoms create an effective barrier and disconnect the two semi-infinite vacancy wires. Such a junction is fundamentally different from break junctions because the tunneling barrier created by the S atoms possesses a detailed electronic structure which contributes to current transmission. The transmission spectrum of the junction created by 2 S atoms (Figure 4) is a convolution of a tunneling current between the atomic wires, tunneling through sulfur states introduced

by the sulfur atoms constituting the junction and interferences between electron waves travelling on the atomic wire and those tunneling through the S junction. The main features of the transmission spectra can however be illustrated with a tight binding model. The atomic junction discussed above provides a platform for connecting a molecule to two atomic wires and measure the conductance of the molecule (Figure 5).

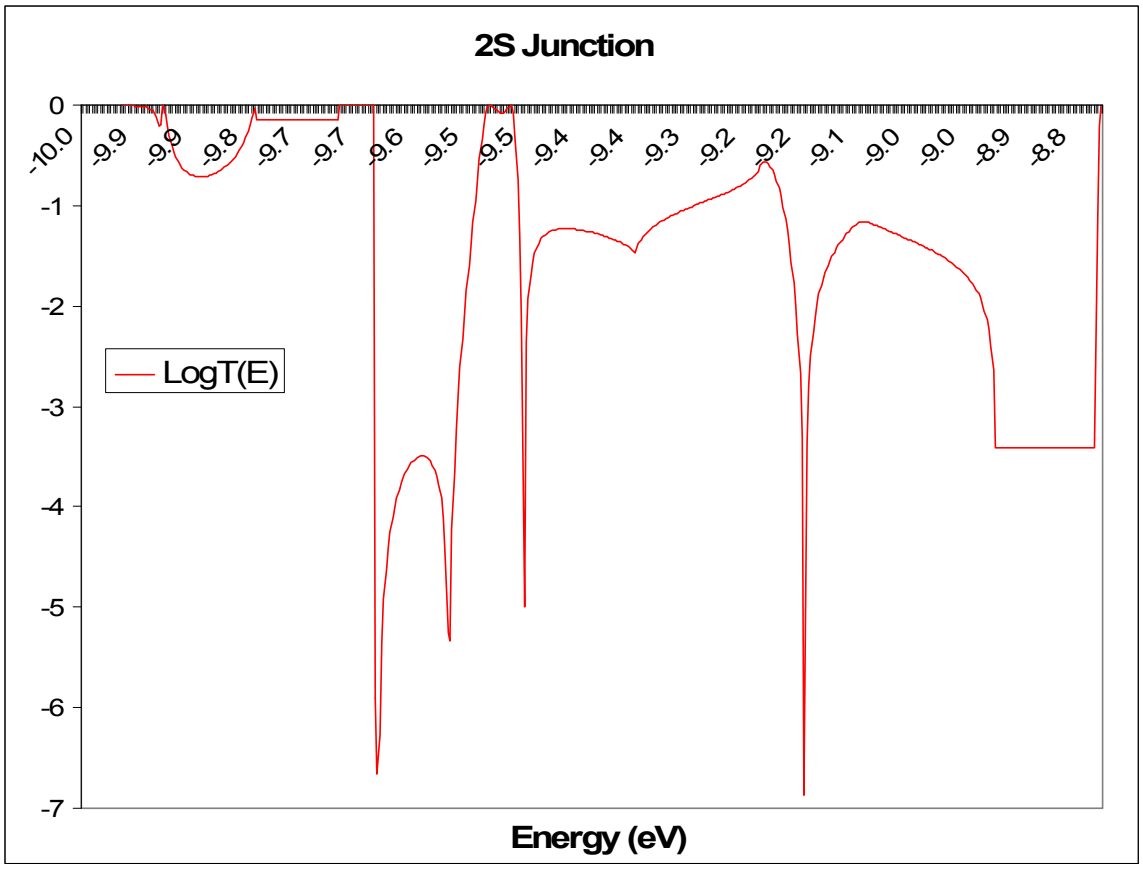

Figure 4: Transmission spectra for an atomic junction created by introducing 2 S atoms in an atomic wire. Sharp interference patterns are observed at well-defined energies.

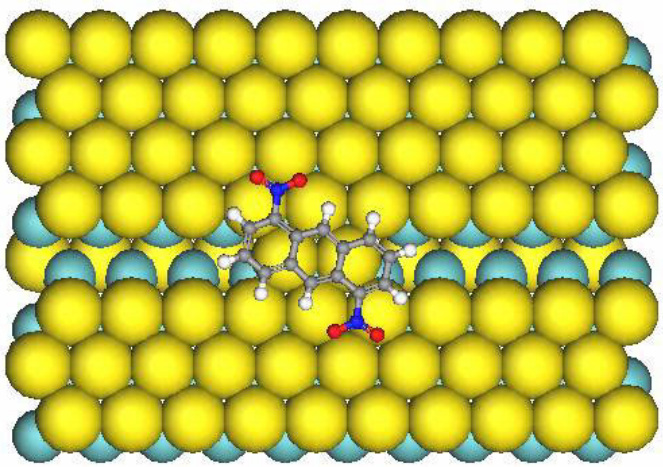

Figure 5: Model of an atomic junction created by insertion of 2 S atoms in an atomic wire with a single molecule logic gate inserted.

Conclusion

In this study it is shown that a pseudo-ballistic atomic wire is created on a MoS_2 surface by extraction of sulfur atoms. Dangling bonds on the surface Mo atoms introduce states in the vicinity of the metal electrodes' Fermi level, which are responsible for the wire's conductance. Furthermore, it is shown that a structured atomic junction of controllable length can be constructed by reintroducing sulfur atoms into the wire's existing vacancies. The atomic wire and the junction offer a framework to study the conductance properties of single molecules.

References

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