Surface-gate-defined single-electron transistor in a MoS$_2$ bilayer

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Abstract

We report the multi-scale modeling and design of a gate-defined single-electron transistor in a MoS$_2$ bilayer. By combining density-functional theory and finite-element analysis, we design a surface gate structure to electrostatically define and tune a quantum dot and its associated tunnel barriers in the MoS$_2$ bilayer. Our approach suggests new pathways for the creation of novel quantum electronic devices in two-dimensional materials.

Keywords: MoS$_2$ bilayer, single-electron transistor, transition-metal dichalcogenides, ab initio, quantum dot, finite-element modeling, transport simulations

(Some figures may appear in colour only in the online journal)

1. Introduction

Novel two-dimensional (2D) materials such as graphene and transition-metal dichalcogenides (TMDCs) have attracted significant interest due to their unique electronic and optical properties [1-3]. The ultrathin geometry and dangling-bond-free interfaces of 2D materials make them good candidates to integrate on various substrates [4].

The incredible range of properties of 2D materials, including high mobility, controlled bandgap and relative ease of fabrication, make them extremely attractive for nanoelectronic applications [5]. 2D materials are actively being explored for potential post-silicon applications, leading to a new era in integrated technology based on layered 2D materials [5-7]. One key factor of 2D materials is the tunability of their bandgaps in the range from insulators to metals via layer thickness, which makes them ideal candidates for photonic applications [8, 9]. Within this context, we have chosen to explore the potential for 2D materials to impact quantum electronics, where their materials properties are also likely to lead to disruptive change.

Among these 2D materials, MoS$_2$ (a TMDC) is promising for quantum electronics; it possesses interesting layer-dependent properties. For example, by decreasing the thickness of MoS$_2$ from bulk to a single layer, its bandgap switches from indirect to direct and increases by more than 0.6 eV, leading to strong photoluminescence from a single layer [2, 10].

The single-electron transistor (SET) is a device where electrons tunnel one by one to and from a small island through tunnel barriers [11]. The tunneling of an electron and the quantization of charge are controlled by gate electrodes. SETs have extensive applications in nano-electronic devices [12]. They have been proposed as a future alternative to conventional CMOS transistors [13], and have been used as nano-scale electrometers [14], capable of measuring sub-electron charge variations to $10^{-6}$ $q_e/\sqrt{\text{Hz}}$. They also have many applications in single-electron logic circuits [15] and single-electron turnstile devices [16].

Many different techniques are used to fabricate SETs based in materials such as Si [17-19], GaAs [20] and carbon nanotubes [21]. There is a significant body of literature on fabricating SETs. For example: Thelander et al have designed gold-nanoparticle SETs using carbon nanotubes as leads at 200 K [22]. Klein et al reported a colloidal chemistry technique to create cadmium-selenide nanocrystals of varying...
sizes [23], and Kim et al used a focused-ion-beam technique to fabricate SETs operating at room temperature [24]. Recently, there has been extensive work to define quantum-dot devices in 2D materials including graphene [25] and TMDCs [26]. Surface gates have been used to create tunable tunnel barriers in Si interfaces [27], but to date there have been no reported designs of electrostatically tunable tunnel barriers and tunable quantum dots in 2D MoS2.

It is known that the MoS2 monolayer band structure shows no response to applied electric fields [28]. In contrast, MoS2 bilayers exhibit significant band-structure modification under perpendicular electric field [28, 29].

Here we present a lithographically appealing design of a surface-gate-defined SET in a MoS2 bilayer, and model its physical characteristics. The structure of our proposed device is a MoS2 bilayer sandwiched between a HfO2 substrate and cap layer, with metallic surface gates, as shown in figure 1(a). We use the surface-gate electric fields to modify the local band structure in the MoS2 bilayer. We present a design of the device obtained through multi-scale modeling. Recently, a more conventional gate design has been used to demonstrate quantum confined structures on a few layers of WSe2 with tunnel barriers defined by electric fields at a temperature of 240 mK [26].

We modeled a MoS2 bilayer using density-functional theory (DFT) to study the effects of electric field on the band structure of MoS2 bilayers. We performed numerical simulations to calculate the potential at the MoS2 bilayer due to the surface gates and used this potential to study the modification of the lowest unoccupied molecular orbital (LUMO) of the MoS2 bilayer. We modeled our device geometry (obtained from the LUMO bending of the MoS2 bilayer) with the commercial finite-element analysis simulation tool COMSOL [30] to obtain the self-capacitance of the SET island and the capacitances between the island and each of the electrodes. We used these capacitances in numerical simulations of transport through the SET at a temperature of 1 K and studied its physical characteristics. We iteratively cycled through COMSOL and numerical simulations to obtain a consistent picture of the device.

This paper is organized as follows: we begin our discussion with the structure of our device geometry followed by DFT modeling of the MoS2 bilayer and the COMSOL modeling. Then we present the influence of the potential due to surface gates in the MoS2 bilayer plane and numerical transport simulations of the resulting SET device.

2. Lithographic structure of our SET device

The most important feature of the gate design shown here, as opposed to more conventional gate-defined quantum dot designs (such as, for example, [26]) is that the creation of potential wells directly underneath and in the near field of the metallic surface gates should allow more-precise lithographic definition of the quantum features in the active layer. In the conventional approach to creating sub-surface quantum dots, the relatively large distance from the gate layer to the active layer gives rise to a complicated potential landscape that must be solved for self-consistently with Schrödinger–Poisson solvers. Conversely, as our active layer is in the near field of the surface gates, the potential should more precisely follow the metallization on the surface layer. This in turn means that more complicated integrated circuits should be realizable, for example multi-island dot structures or integrated qubit/read-out type circuits. This design ethos is exemplified by the surface gate defined SETs induced at a silicon/silica interface by Angus et al [27].
Our SET device has an MoS$_2$ bilayer as an active layer sandwiched between HfO$_2$ insulating layers as shown in figure 1(a). The top metallization layer comprises four gates G1–G4. Gates G1 and G2 of figure 1(a) are approximated as infinitely long lines of charge, the intersection of which defines our island via the doubly strong steep potential in the active layer at depth (MoS$_2$ bilayer). To establish source and drain leads in the active layer at depth, we use gates G3 and G4 to create similarly strong potential landscapes directly underneath gate G1. These source and drain leads are separately connected to external voltage controllers, enabling direct modification of their chemical potential(s), $\mu$ and bias, $V_{bd}$.

For convenience, gates G3 and G4 are approximated as one infinite line of charge with a missing, finite segment centered on the intersection of gates G1 and G2. For our treatment, this dictates $V_3 = V_4$, but that condition is not necessary if the semi-infinite gates G3 and G4 are treated separately and identical source and drain behavior is not required. It should also be noted that $V_3 \approx V_4 = V_2$ achieves comparable island and lead depths below the barrier peaks; $V_3 = V_4 = V_2$ is not a constraint. Gate G1 is therefore effectively a plunger gate controlling the depth of all components, which could in principle be operated in isolation from the rest (although $V_1 + V_3 > V_2$ is required to avoid an unintended perpendicular lead under gate G2). For simplicity here, however, we set the general gate voltage as $V_1 = V_2 = V_3 = V_4 = V_5$.

3. DFT modeling of molybdenum-disulphide bilayer structures

Although the effects of electric field on the band structure of MoS$_2$ bilayers are available in the literature, the focus has either been on behavior over extreme field strength ranges (with sampling too coarse for our device) [28] or on the bandgap response without specific discussion of the LUMO physics [29]. Thus, to achieve the insight necessary to design our device, we explore the LUMO physics directly in the target operating regime.

We investigated the band structure of an optimized, $A$–$A'$ stacked (Mo atoms in the top monolayer above the S atoms of the bottom monolayer), 2H-phase MoS$_2$ bilayer for varying electric fields. $A$–$A'$ is the most-reported stacking order for bulk MoS$_2$ and it is very close in energy to the most stable stacking order $A$–$B$ [29]. A perspective view of the MoS$_2$ bilayer is shown in figure 1(b).

To model the effects of electric field on the MoS$_2$ bilayers, we used DFT in CRYSTAL09 [31, 32]. MoS$_2$ has hexagonal symmetry and belongs to the $P6_3/mmc$ space group, with lattice parameters $a = 3.17$ Å and $c = 12.324$ Å [33]. We created the bilayer unit slab by cutting a (001) plane from a bulk MoS$_2$ model, and including vacuum to a total cell height of $c = 500$ Å. The exchange and correlation terms were described via the PBEsol functional [34], which generally predicts lattice constants more accurately than PBE and LSDA (thus improving the equilibrium properties of solids), and also handles the electronic response to potentials better than most GGA functionals [34]. We used Gaussian basis sets; Mo_SC_HAYWSC311(d31)G_cora_1997 [35] for Mo atoms (a Hay-Wadt effective-core pseudopotential [36] combined with a valence-electron basis set) and S_pob_TZVP_2012 [37] for S atoms (an all-electron basis set). We set a $8 \times 16 \times 1$ Monkhorst–Pack [38] $k$-point mesh. We have ignored the effect of spin–orbit coupling (SOC) in our calculations, as SOC has been shown to not cause any splitting in the conduction band minima of MoS$_2$ bilayers [39, 40].

We optimized the geometry of a MoS$_2$ bilayer unit slab under zero electric field and then used this optimized geometry to study the effects of electric field on the band structure of MoS$_2$ bilayers. We calculated the band structure along the high-symmetry path $\Gamma$–M–K–$\Gamma$. Band structures were calculated for varying electric fields applied perpendicular to
and HOMO under electric field. We excluded these points from further calculations as their band structures are qualitatively different and we believe that these inconsistent points do not represent the underlying physics.

We applied a linear fit to the filtered LUMO and HOMO data, and set zero energy at the LUMO intercept (at zero field). As an electric field is applied up to $\pm 0.2 \text{ V \AA}^{-1}$, the LUMO bends through 138 meV. Thus electron confinement is achievable in a MoS$_2$ bilayer via electric-field modification of the LUMO, which occurs at a rate of 690 meV ($\text{V \AA}^{-1}$)$^{-1}$.

4. Circuit simulations in COMSOL

We modeled our device geometry in COMSOL [30] to get the capacitances between all the device components. The geometry (obtained through potential simulations discussed later and converged by iteratively cycling through numerical potential and transport simulations and COMSOL) used for COMSOL modeling is an island of diameter 12 nm and the same thickness as the MoS$_2$ bilayer. There are 16 nm wide MoS$_2$ barriers between the island and in-plane source and drain. The surface gates are modeled as two intersecting wires each of 5 nm radius sitting atop the 7 nm HfO$_2$ cap layer. We excluded gates G3 and G4 of figure 1(a) from our COMSOL modeling as they are collinear with gate G1 and do not contribute significantly to the total capacitance of the island.

The ground plane is 11 nm (thickness of the HfO$_2$ substrate) below the island. The capacitances were calculated following the COMSOL AC/DC module user manual [41].

For an island of diameter 12 nm, the self-capacitance is $C_{i\Sigma} = 8.45 \text{ aF}$ which corresponds to a charging energy of $q_e^2/C_{i\Sigma} = 18.9 \text{ meV}$, where $C_{i\Sigma}$ is defined as

$$C_{i\Sigma} = C_{is} + C_{id} + C_{G1} + C_{G2} + C_{isp}$$

Here $C_{is} = 0.252 \text{ aF}$ is the capacitance between the island and the buried source, $C_{id} = 0.253 \text{ aF}$ is the capacitance between the island and the buried drain, $C_{G1} = 2.2 \text{ aF}$ is the capacitance between the island and gate G1, $C_{G2} = 2.38 \text{ aF}$ is the capacitance between the island and gate G2, and $C_{isp} = 3.35 \text{ aF}$ is the capacitance between the island and the ground plane.

5. Single-electron transport simulations

In our proposed MoS$_2$ SET design, we stress that the surface gates define and electrostatically control the essential elements of the SET, namely the source, island and drain, which are all in the MoS$_2$ bilayer. There are two longer surface gates G1 and G2, perpendicular to each other. Two shorter gates, G3 and G4, are collinear with the G1 gate as shown in figure 1(a). All of these surface gates are electrostatically insulated from each other; this can be done by thermally growing a thin oxide layer between them. The equivalent electrical circuit diagram is shown in figure 1(c), where $R(V_g)$ is the tunnel resistance between the island and the source/
We modeled our device design through in-house numerical simulations. Gates G3 and G4 were placed on top of gate G1, with their finite edges 32 nm apart from each other. The thicknesses of the cap layer and the substrate were 7 nm and 11 nm respectively. A varying DC potential of 0.2–0.34 V was applied to all surface gates. All of these parameters and the DC potential range were finalized through iterations between the COMSOL modeling and the numerical simulations. The top view of the potential created in the MoS2 bilayer plane due to the surface gates is shown in figure 1(d). The intersection of gates G1 and G2 generates a well-defined island in the MoS2 bilayer plane. Gates G1, G3, and G4 define the source/drain wiring configuration and tunable tunnel barriers in the MoS2 bilayer plane. An analytical expression for the potential created due to one of the surface gates is given in the appendix.

Figure 1(e) shows a slice of the potential on top of the MoS2 bilayer plane and underneath gates G1, G3, and G4. Note that there are small local minima in the potential (and the electric field) outside the barriers. The potential difference across the MoS2 bilayer is proportional to the electric field (figure 3(b)). The maximum electric field created across the MoS2 bilayer (for \( V_g = 0.34 \) V) is approximately 4.6 mV Å\(^{-1}\) which is comparable to fields applied across MoS2 bilayer devices in [42], and well exceeded by the breakdown field strength of HfO\(_2\) [43]. We used the linear fit to figure 3(a) to obtain the LUMO bending at fields shown in figure 3(b). This LUMO bending as a function of the position across the MoS2 bilayer plane is shown in figure 3(b). The constraints of achieving a useful device and consequent constraints on the geometry lead us to place the Fermi level at 0.15 meV. This placement of the Fermi level corresponds to a 12 nm diameter island and 16 nm wide tunnel barriers in the MoS2 bilayer plane. The dimensions of the SET are comparable with experimental structures in GaAs/AlGaAs [44]. The size of the island and the width of the tunnel barriers can be controlled through the substrate and the cap layer thicknesses. The LUMO bending is used as an input to numerical transport simulation.

Increases to the general gate voltage, \( V_g \), cause the barrier height to rise, reducing the transmission coefficient. Figure 4 shows the LUMO bending for several values of \( V_g \), together with constant region boundaries. Figure 5(a) shows the variation of tunnel resistance with \( V_g \). While the transmission coefficient decreases with higher \( V_g \), this effect is minimal due to the small barrier height and width. The dominant components of the resistance are the densities of states of the source/drain and the island, which both increase considerably with higher \( V_g \) and the constraint of modifying \( \mu \) to maintain the device geometry. (For details of the tunnel resistance and transport calculations, see the appendix.)

The behavior of the SET can be observed by varying \( V_g \). Figure 5(b) shows the characteristic current passing through the SET at a temperature of 1 K and a bias voltage of 100 \( \mu \)V.

![Figure 4.](image1) LUMO bending as function of position along the source-island-drain axis for varying gate voltages within the device target operating regime. The barrier function behaves linearly with the general gate voltage, \( V_g \). Vertical marks show the boundaries of the labeled regions (black, dashed lines with arrow heads). \( \varepsilon \) is not shown since it varies with \( V_g \), but its value for each \( V_g \) can be identified as the intersections of the corresponding LUMO data traces with the region boundaries.

![Figure 5.](image2) Transport characteristics of the SET device: (a) variation of the barrier tunnel resistance with the general gate voltage, \( V_g \); (b) Characteristic Coulomb blockade peaks for a source/drain bias voltage of 100 \( \mu \)V and at a temperature of 1 K, showing a variable current through the MoS2 bilayer SET.
By varying the surface gate voltage, the overall potential landscape through the MoS$_2$ bilayer changes, thus modifying the barrier heights and the alignment between incoming electron energy and island states. We explicitly consider the high charging energy case, when the classical occupancy of the SET island can change by at most one electron at a time. Hence, effects due to the addition energy have negligible effects upon this SET. When the energy of an incoming electron resonates with the unoccupied energy level in the island, that electron tunnels to the island and current flows through the SET which is shown by the peaks in figure 5(b). On the other hand, when none of the island states are available to the incoming electron, it cannot tunnel to the island and the current through the SET drops to zero, defining the Coulomb-blockade region between the peaks. The height variation between the current peaks arises from the changing response of the MoS$_2$-bilayer density of states in the source/drain and island, in contrast to an ideal metallic SET.

6. Conclusions

The surface gate approach to quantum devices is quite flexible and amenable to scalable integrated devices. With identical gate voltages, we already have significant control over barrier height, the energy states in the island and hence, electron tunneling. One could achieve further independent control of the incoming electron energy or island states by applying different voltages to each gate. Moreover, due to the small size of the island, our preliminary modeling holds promise for relatively high-temperature single-electron devices.

The electric-field-based tunability of the HOMO and the LUMO of the MoS$_2$ bilayers opens possibilities for the surface gate approach to be used for potential future opto-electronic devices. For example, one could consider electrically controlling the band gap to create electrically tunable emission from quantum dot regions, where both the quantum dot geometry and emission wavelength are controlled by the surface-gate potentials.

Here we have studied an embedded MoS$_2$ bilayer system. However, we would expect our results to be easily applicable to other bilayer 2D materials that exhibit a LUMO response to electric field (e.g., MoSe$_2$, MoTe$_2$ and WS$_2$ [28]), with only minimal modification of the applied surface potentials.

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Appendix

Here we present a brief overview of the methods and techniques used to design and model the MoS$_2$ bilayer SET. We show our analytical and/or numerical treatment of the potential across the bilayer plane, capacitance of the device, and details of our calculations of the resistance of (and current through) the SET.

Defining $x$ as the distance transverse to gate G1 in the bilayer plane (with zero value directly underneath G1), $y$ similarly with reference to G2, and $z$ as the bilayer-ground plane separation, the electric potential difference created across the MoS$_2$ bilayers is the summed effect from all gates, which is analytic. For example, the potential due to gate G1 is [45]

$$V = \frac{\lambda}{2\pi\epsilon_0\epsilon_r} \log \left[ \frac{x^2 + (z - d)^2}{x^2 + (z + d)^2} \right],$$  \hspace{1cm} (A1)

where $\lambda$ is the linear charge density of gate G1, $\epsilon_r$ is the dielectric constant of HfO$_2$ [46] and $d$ is the distance of gate G1 from the ground plane. The other potentials are similarly derived. The finite difference between the top and bottom edges of the bilayer is evaluated to provide the field strength across the bilayer.

Having obtained a 2D map of the potential difference across the bilayer plane—e.g., figure 1(d) in the main paper—we then consider its effect on the MoS$_2$ band structure at fields from 0 to 0.2 V Å$^{-1}$ in steps no larger than 0.01 V Å$^{-1}$ and smaller as required to verify linear behavior.

We are particularly interested in a minimal substrate footprint, and the constraint of reasonable applied field strength. Therefore in the main paper, without loss of generality in the method, we explicitly consider the high charging energy case, when the classical occupancy of the SET island can change by at most one electron at a time. Due to the small barrier height of our device, there is only ever at most one unoccupied level on the island. Higher lying unoccupied levels would exceed the barrier height, and hence be unbound. Thus, the extent of the LUMO bending ($\Delta L$, measured from the left asymptotic value to the peak), created must match well to the charging energy of the system [47]:

$$\Delta L \approx E = \frac{1}{2} Q^T C^{-1} Q,$$  \hspace{1cm} (A2)

where $Q$ is the charge on the SET, defined by $Q = [Q_s, Q_G, Q_{G1}, Q_{G2}, Q_u, Q_{gp}]^T$. $C$ is the capacitance matrix between all the device components and is defined as:

$$C = \begin{pmatrix}
C_s \Sigma & -C_{is} & -C_{G1} & -C_{G2} & -C_d & -C_{gp} \\
-C_{is} & C_i \Sigma & -C_{G1} & -C_{G2} & -C_d & -C_{gp} \\
-C_{G1} & -C_{G1s} & C_{G1} & -C_{G2} & -C_d & -C_{gp} \\
-C_{G2} & -C_{G2s} & -C_{G2} & C_{G2} & -C_d & -C_{gp} \\
-C_d & -C_{ds} & -C_{G1} & -C_{G2} & C_{\Sigma} & -C_{4p} \\
-C_{gp} & -C_{gps} & -C_{gpG1} & -C_{gpG2} & -C_{gpC} & C_{\Sigma Gp}
\end{pmatrix}$$  \hspace{1cm} (A3)
the terms of which are obtained via COMSOL modeling as described in the main paper. The boundary conditions used follow [41].

The tunnel resistance cannot be evaluated exactly. The usual approach to an arbitrary barrier, the WKB approximation, is not directly applicable if the incoming energy of the electron matches the barrier energy at any point. In fact, not only does the approximation not hold at those points, but also in their vicinities. The connection formulas, using an Airy function ansatz to patch difficult regions, are the standard way to circumvent this issue; however, the conditions for their use are not met by this barrier. Accordingly, we make the approximation that the barrier is finite and rectangular, with the identical width (16 nm) and height equal to the peak value of LUMO bending, called \( L_{\text{peak}} \) while setting \( L(-200) = 0 \). Since such a barrier is definitely larger, and no thinner, than the original barrier, its transmission coefficient is guaranteed to be lower than any experimental device. The transmission coefficient of a finite square barrier is [48]:

\[
T = e^{-2\gamma},
\]

where

\[
\gamma = \frac{1}{\hbar} \int_{0}^{a} \sqrt{2m(L_{\text{peak}} - \varepsilon)} \, d\varepsilon = \frac{a}{\hbar} \sqrt{2m(L_{\text{peak}} - \varepsilon)},
\]

(\( \hbar = \hbar/2\pi \) is the reduced Planck’s constant, \( m = 0.38 m_e \) is the effective mass of electrons in the MoS\(_2\) bilayer [49], \( a \) is the width of the tunnel barrier, and \( \varepsilon \) is the energy of an incoming electron in the MoS\(_2\) bilayer.

Here \( \varepsilon \) can also be regarded as \( \varepsilon = V_{sd} + E_{th} + \mu \), where \( V_{sd} \) is the source-drain bias, \( E_{th} \) is the thermal energy described by the Fermi smearing incorporated below, and \( \mu \) is the chemical potential of the electron which is set by the absolute value of each in tandem has no effect on \( V_{sd} \). We assume that the incoming electron has \( \mu \) such that \( \varepsilon \approx 0.38[L_{\text{peak}} - L(-200)] = 0.38L_{\text{peak}} \); this defines the points where \( L = \varepsilon \) (the region boundaries) at consistent locations in space—enforcing the desired geometry with large charging energy. The left edge of \( L \) has been set as zero energy; the absolute value of \( \mu \) is somewhat different to the \( \varepsilon \) marked by the dashed line in figure 3(b). Figure 4 shows \( L(x) \) for several values of \( V_g \), together with the constant region boundaries. In principle, this assumption could be relaxed as needed to describe a particular physical device.

As an example, figure 3(b) in the main text shows the particular case of the optimized structure under \( V_g = 0.27 \) V, where \( \mu \) is set such that \( \varepsilon \) is equal to the black-dashed line. There, the island has 12 nm diameter and 16 nm wide tunnel barriers. \( L \) is the barrier height created by LUMO bending due to general gate voltage \( V_g \).

The barrier tunnel resistance is given by [50]

\[
R_T^{-1} = \frac{2\pi q_e^2}{\hbar} |T(\varepsilon)|^2,
\]

where \( |T(\varepsilon)| \) is the transmission coefficient, \( \rho_1 \) is the density of states in the source/drain electrodes and \( \rho_2 \) is the density of states in the island. We approximate the density of states by assuming constant band bending over the leads (and island), giving:

\[
\rho = \int_{\varepsilon_0}^{\varepsilon_f} D(\varepsilon) \, d\varepsilon = D(\varepsilon - \varepsilon_0) \times (\varepsilon - \varepsilon_0);
\]

(\( \varepsilon_f \) is the Fermi level and \( \varepsilon_0 \) is the energy of the lowest level in the source/drain leads of MoS\(_2\) bilayer plane. This corresponds to making the assumption that electron-hole recombination is negligibly slow compared to the tunneling rate.) \( D(\varepsilon) \) is the density of states per unit energy in a 2D solid, given by

\[
D(\varepsilon) = \frac{4\pi m}{\hbar^2} \times A,
\]

where \( A \) is the area. Hence,

\[
\rho_1 = \frac{4\pi m}{\hbar^2} (\varepsilon - \varepsilon_0) \times A, \quad \text{for} \quad \varepsilon > \varepsilon_0, \quad \text{and}
\]

\[
\rho_2 = \frac{4\pi m}{\hbar^2} (\varepsilon - \varepsilon_0) \times A, \quad \text{for} \quad \varepsilon > \varepsilon_0.
\]

Here, \( A_s \) and \( A_l \) are the areas for the source and island respectively. The island and lead in-plane areas were estimated as \( \pi (6 \text{ nm})^2 \) and \( 400 \times 20 \text{ nm}^2 \), respectively which leads to tunnel resistances of 270 to 360 k\( \Omega \) for 0.2 V \( \leq V_g \leq 0.34 \) V, using this idealized design in perfect bilayer MoS\(_2\) in HFO\(_2\). Practically, many factors involved in experiment (e.g., fabrication imprecision, defects, etc.) often accumulate to affect resistances by several orders of magnitude.

Although we use a semiconductor material (MoS\(_2\) bilayer) in the design of our SET, after connecting it with the external batteries the Fermi level rises into the conduction band in the source/island/drain regions while sitting below the conduction band in the tunnel barrier regions. Thus, the SET behaves effectively like a metal-insulator-metal junction and all the standard approximations for metallic SETs are still applicable to our SET.

To determine the current through the SET, we need the tunneling rates on and off the island for different charge configurations of the SET. These tunneling rates are given by [12]

\[
\Gamma_{\chi} = \frac{1}{q_e^2 R_T} \exp\left(\frac{\Delta E_{\chi}^n}{k_B T}\right) - 1,
\]

(\( \chi \) denotes the source or drain and \( \chi \) the island. \( \Delta E_{\chi}^n \) is the energy difference between different charge configurations of the SET and is given by

\[
\Delta E_{\chi}^n = E(n - 1) - E(n) + V_q q_e
\]

Here \( V_q \) is the chemical potential applied to the source/drain lead.

The current through the SET is sum of the probabilities for all possible tunneling rates on and off the island. This current is given by

\[
I = q_e \sum_{\chi = \text{sd}} \sum_{n = -\infty}^{\infty} \rho_1 \Gamma_{\chi}^n - \Gamma_{\chi}^n,
\]
where \( p_n \) is the probability that island is in a state with \( n \) excess electrons. These probabilities are calculated by the master equation as described in [47].

References

[1] Novoselov K S, Geim A K, Morozov S V, Jiang D, Katsnelson M I, Grigorieva I V, Dubonos S V and Firsov A A 2005 Nature 438 197
[2] Mak K F, Lee C, Hone J, Shan J and Heinz T F 2010 Phys. Rev. Lett. 105 136805
[3] Fiori G, Bonaccorso F, Iannaccone G, Palacios T, Neumaier D, Sebaugha A, Banerjee S K and Colombo L 2014 Nat. Nanotechnol. 9 768
[4] Bolotin K I, Sikes K J, Jiang D, Klima M, Fudenberg G, Hone J, Kim P and Stormer H L 2008 Solid State Commun. 146 361
[5] Wang H, Yu L, Lee Y-H, Shi Y, Hsu A, Chin M L, Li J-J, Dubey M, Kong J and Palacios T 2012 Nano Lett. 12 4674
[6] Akinwande D, Petrone N and Hone J 2014 Nat. Commun. 5 5678
[7] Qian X, Wang Y, Li W, Lu J and Li J 2015 2D Mater 2 032003
[8] Ponraj J S et al 2016 Nanotechnology 27 462001
[9] Wang Q H, Kalantar-Zadeh K, Kis A, Coleman J N and Strano M S 2012 Nat. Nanotechnol. 7 699
[10] Speddingi A, Sun L, Zhang Y, Li T, Kim J, Chim C Y, Galii G and Wang F 2010 Nano Lett. 10 1271
[11] Singh V P, Agrawal A and Singh S B 2012 Int. J. Soft Comput. Eng. 2 502 (http://citeserix.ist.psu.edu/viewdoc/download?doi=10.1.1.460.1729&rep=rep1&type=pdf)
[12] Grabert H and Devoret M H 1992 Single charge tunneling: Coulomb blockade phenomena in nanostructures (New York: Plenum Press)
[13] Mahapatra S, Vaish V, Wasshuber C, Banerjee K and Ionescu A M 2004 IEEE Trans. Electron Devices 51 1772
[14] Devoret M and Schoelkopf R 2000 Nature 406 1039
[15] Kim S J, Lee J J, Kang H J, Choi J B, Yu Y-S, Takahashi Y and Hasko D G 2012 Appl. Phys. Lett. 101 183101
[16] Yuan Z L et al 2002 Science 295 102
[17] Fuechsle M, Mahapatra S, Zwanenburg F A, Friesen M, Eriksson M A and Simmons M Y 2010 Nat. Nanotechnology 5 502
[18] Hofheinz M, Jehl X, Sanquer M, Molas G, Vinet M and Debelius S 2006 Appl. Phys. Lett. 89 143504
[19] Shin S J, Jung C S, Park B J, Yoon T K, Lee J J, Kim S J, Choi J B, Takahashi Y and Hasko D G 2010 Appl. Phys. Lett. 97 103101
[20] Knobel R G and Cleland A N 2003 Nature 424 291
[21] Stokes P and Khondaker S I 2008 Appl. Phys. Lett. 92 262107
[22] Thelander C, Magnusson M H, Deppert K, Samuelson L, Poulsen P R, Nygard J and Borggreen J 2001 Appl. Phys. Lett. 79 2106
[23] Klein D L, Roth R, Lim A K L, Alivisatos A P and McEuen P L 1997 Nature 389 699
[24] Kim T W, Choo D C, Shim J H and Kang S O 2002 Appl. Phys. Lett. 80 2168
[25] Stampfer C, Schurtenberger E, Molitor F, Güttinger J, Ihn T and Ensslin K 2008 Nano Lett. 8 2378
[26] Song X-X et al 2015 Nanoscale 7 16867
[27] Angus S J, Ferguson A J, Dzurak A S and Clark R G 2007 Nano Lett. 7 2051
[28] Ramasubramaniam A, Naveh D and Towe E 2011 Phys. Rev. B 84 205325
[29] Liu Q, Li L, Li Y, Gao Z, Chen Z and Lu J 2012 J. Phys. Chem. C 116 21556
[30] COMSOL MULTIPHYSICS® 2008 (Burlington, MA: COMSOL, Inc.) version 5.1
[31] Dovesi R, Orlando R, Civallieri B, Roetti C, Saunders V R and Zicovich-Wilson C M 2005 Z. Kristallogr. 220 571
[32] Dovesi R et al 2009 Crystal09 User’s Manual (Torino: University of Torino)
[33] Dickinson R G and Pauling L 1923 J. Am. Chem. Soc. 45 1466
[34] Perdew J P, Ruzsinszky A, Csonka G I, Vydrov O A, Scuseria G E, Constantin L A, Zhou X and Burke K 2008 Phys. Rev. Lett. 100 136406
[35] Corã F, Patel A, Harrison N M, Roetti C and Catlow C R A 1997 J. Mater. Chem. 7 959
[36] Ehlers A, Böhme M, Dapprich S, Göbbi A, Höllwarth A, Jonas V, Köhler K, Stegmann R, Veldkamp A and Frenking G 1993 Chem. Phys. Lett. 208 111
[37] Peintinger M F, Oliveira D V and Bredow T 2013 J. Comput. Chem. 34 451
[38] Monkhorst H J and Pack J D 1976 Phys. Rev. B 13 5188
[39] Fan X, Singh D J and Zheng W 2016 J. Phys. Chem. Lett. 7 2175
[40] Dou X, Ding K, Jiang D, Fan X and Sun B 2016 ACS Nano 10 1619
[41] COMSOL 2013 AC/DC User’s Guide 4th edn (Burlington, MA: COMSOL, Inc.)
[42] Xie X, Sarkar D, Liu W, Kang J, Marinov O, Deen M J and Banerjee K 2014 ACS Nano 8 5633
[43] Sire C, Blondowski S, Gordon M J and Baron T 2007 Appl. Phys. Lett. 91 242905
[44] van der Wiel W G, De Franceschi S, Elzerman J M, Fujisawa T, Tarucha S and Kouwenhoven L P 2002 Rev. Mod. Phys. 75 1
[45] Jong L M, Greenlee A D, Conrad V I, Hollenberg L C and Jamieson D N 2009 Nanotechnology 20 405402
[46] Jones M, Kwon Y and Norton D 2005 Appl. Phys. A 81 285
[47] Conrad V I, Greentree A D, Jamieson D N and Koppinen P 2003 Bias and temperature dependence analysis of the tunneling current of normal metal-insulator-normal metal tunnel junctions Master’s Thesis University of Jyväskyla Department of Physics