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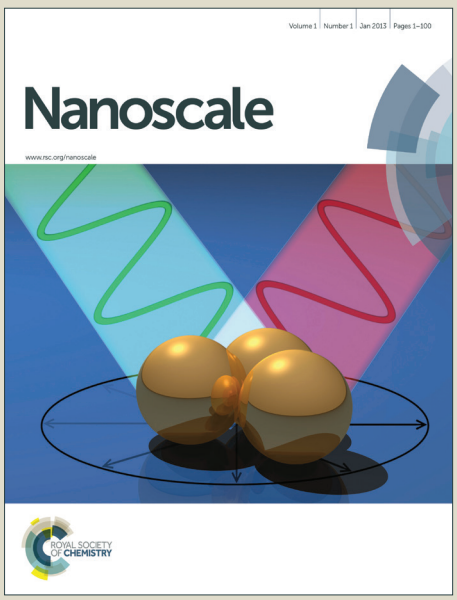

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Gate Controlled Molecular Switch Based on picene-F_4TCNQ Charge-Transfer Material

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We show that the recently synthesized charge-transfer material picene-F_4TCNQ can be used as a gate-voltage controlled molecular switch. The picene-F_4TCNQ system is compared with the extensive characterized anthraquinone-based molecular system, which is known to exhibit large switching ratios due to quantum interference effects. In case of picene-F_4TCNQ we find switching ratios larger by one order of magnitude. Further, our calculations reveal that the picene-F_4TCNQ system resembles remarkable well the I-V characteristics of a classical diode. The reverse-bias current of this molecular diode can be increased two orders of magnitude by an external gate voltage. Based on density-functional theory calculations we show that the hybrid states formed by the picene-F_4TCNQ system are playing the key role to determine the transport properties. We further conclude that the tuning of quantum transport properties through hybrid states is a general concept which opens a new route towards functional materials for molecular electronics.

1 Introduction

The basic idea of molecular electronics is that typical functionality needed to build integrated circuits has to be realized by single molecules. This idea was already part of the pioneering theoretical description of a molecular diode given by Ratner and coworkers many years ago[1]. Since then it was repeatedly discussed[2–4] why it is not easy to actually build a satisfying molecular version of the classical semiconductor diode. In practice we face the problem that in nearly all molecular devices the I-V characteristics is rather symmetrical even when using a highly asymmetric molecule. As a result the attempts to manufacture molecular diodes or transistors resulted in rather poor rectification or switching ratios compared to conventional semiconductor devices[5]. The criteria for achieving diode-like high rectification ratios in molecular junctions were already suggested by Ratner et al.[1]. Their proposal was that organic charge-transfer materials where two molecular subunits each carry an opposite charge should be suitable for the task. However, from this theoretical work it was also concluded that strong coupling between the molecular subunits may completely screen the desired effect.

Recently this idea was picked up by[6] and they presented rectification ratios $R = 2 \ldots 3$. In this paper we present theoretical results on the charge transfer material picene-F_4TCNQ[7]. This material was synthesized very recently for the first time and the experiments indicated new electronic states due to the charge transfer. These electronic levels close to the Fermi energy are beneficial for transport and the additional flat geometry together with the weak π-π coupling of the molecular building blocks are a perfect match for the requirements given by Ratner.

We will show that this system can reach much higher rectification ratios up to $R \approx 20$ which is at least one order of magnitude higher than other values reported before. Our system also exceeds the recently published results by Batra and coworkers in terms of achievable values for R[11]. We compare our theoretical results to calculations on an anthraquinone based molecular switch. The anthraquinone system was chosen because it is well characterized and known to perform as an molecular switch[8–10]. Furthermore theoretical calculations with different contact materials and reliable measurements are available for comparison with our results[12–15]. We also investigate the dependence of the I-V characteristics on an external gate voltage. Apart from electrochemical gating[16] or redox active switching[12] this seems to be the most promising way to manufacture working active electronic devices.

Results and Discussion

The material under consideraion here is built from picene and fluorinated TCNQ molecules forming molecular crystals or films with an 1:1 composition which can be synthesized by co-evaporation[7]. The main building block in the crystal is a dimer like structure of picene and TCNQ with flat orientation with respect to each other. Therefore, we start our discussion with a short comparison of the electronic structure of the free

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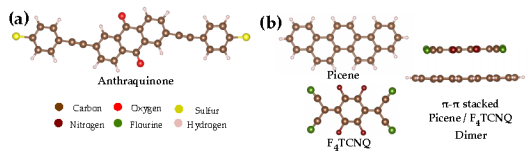

Fig. 1 Schematic drawings of the molecules as used in the electronic structure and quantum transport calculations: (a) the anthraquinone derivate and (b) the picene-F_4TCNQ -dimer.

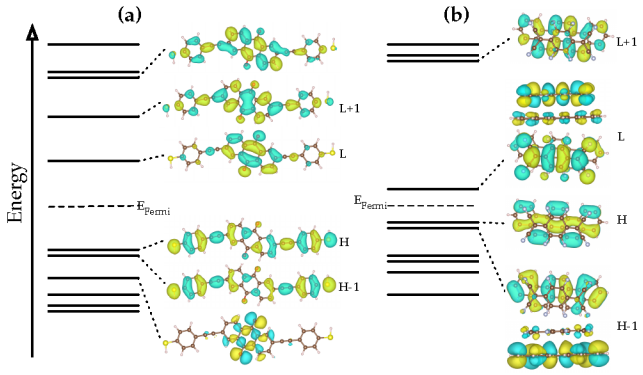

Fig. 2 Electronic structure close to the Fermi level as obtained from density functional theory calculations for (a) the anthraquinone derivate and (b) the picene-F_4TCNQ -dimer.

picene-F_4TCNQ -dimer and anthraquinone derivate based on density functional theory (DFT) calculations. The structures of the investigated molecules are shown in Fig. 1.

The electronic structure of the anthraquinone derivate as depicted in Fig. 2 a) has already been discussed in detail elsewhere [10,14]. It shows the typical properties of a π-conjugated molecular material (semiconductor) having a HOMO - LUMO gap of about 1.6 eV and an almost symmetric level arrangement. The HOMO and HOMO-1 levels are delocalized and span the whole molecule including the thiol anchor units. The sprawling side structures guarantee that electron withdrawing or pushing effects of the anchor units modify only slightly the underlying electronic structure of the anthraquinone core.

The electronic structure of the picene-F_4TCNQ system on the other hand is determined by the occurrence of hybrid orbitals which are formed between the π-conjugated picene and the F_4TCNQ acceptor. The HOMO-1 and LUMO orbital of picene-F_4TCNQ are formed from states of the free picene and F_4TCNQ. This hybridization induces a charge transfer of about 0.2 e from the picene to the F_4TCNQ [7].

Based on the electronic structure calculations the transport properties have been obtained using the non-equilibrium Green function formalism (NEGF). Both molecular systems are sandwiched between two Au(111) leads. Fig. 3 shows the I-V curves without gate voltage (black lines) for the a) anthraquinone and b) picene-F_4TCNQ system. Both curves show typical details which are expected from the I-V-characteristics of a molecular junction. Due to the weak coupling of both molecular systems to the Au(111) leads both systems show features which can be attributed to distinct molecular orbitals.

In a very simplified picture if bias voltage is rising then more orbitals will contribute to the conduction through the junction and the current increases. Peaks or regions of negative differential resistance (NDR) occur if distinct levels contribute to the conduction on low bias and do not contribute in the case of high bias voltages due to e.g. the lowering of the coupling strength between the molecular orbital and the lead [12,17]. Such a situation can be seen for example in the anthraquinone system at ≈ 0.2 V and ≈ 0.5 V bias voltage and for the picene-F_4TCNQ system at $V_{bias} \approx 0.25$ V.

The effect of increasing bias voltage on the electron transmission spectra of the two molecular junctions is depicted in Fig. 3 c) and d) respectively. In the zero bias transmission function the picene-F_4TCNQ -system (solid line) has a prominent feature at 0.2 eV above the Fermi level (E_F) which can be attributed to the LUMO level of the dimer. The features right below E_F are therefore linked to the HOMO, HOMO-1 etc. dimer orbitals. Above 0.3 eV picene-F_4TCNQ exhibits no features relevant to transport.

The main effects of an applied bias voltage to a molecular junction are (i) shifting the transmission spectra with respect to V_{bias}, (ii) strengthening and dampening of transmission features due to the bias induced changes in the molecule-lead coupling and (iii) widening of the energy window in which transmission peaks contribute to the current. Therefore if we apply a positive bias to the picene-F_4TCNQ junction we are shifting transmission features which correspond to occupied HOMO-n orbitals into the energy window relevant for conduction. For negative bias the transmission peaks originating from the HOMO and LUMO are shifted out of that window and due to the large energy gap between the LUMO and LUMO+n levels (see Fig. 2b) there are no additional levels which can contribute to the conduction. Only in the case of very high bias voltages > 1 V additional transmission features can appear and the current starts to rise also for negative bias.

For the anthraquinone junction the situation is completely different. We observe a somewhat lower density of transmission peaks for energies above E_F (see Fig. 3c). The changes in the bias voltage do not significantly change the overall density of transmission peaks in the energy window around E_F contributing to the conduction. Hence the absolute value of the current through the junction is approximately the same whether we apply a negative or positive bias voltage. Our results are in good agreement with other theoretical estimates [12,13]. In the picene-F_4TCNQ junction (Fig. 3b) the current for the negative bias case stays almost at zero up to

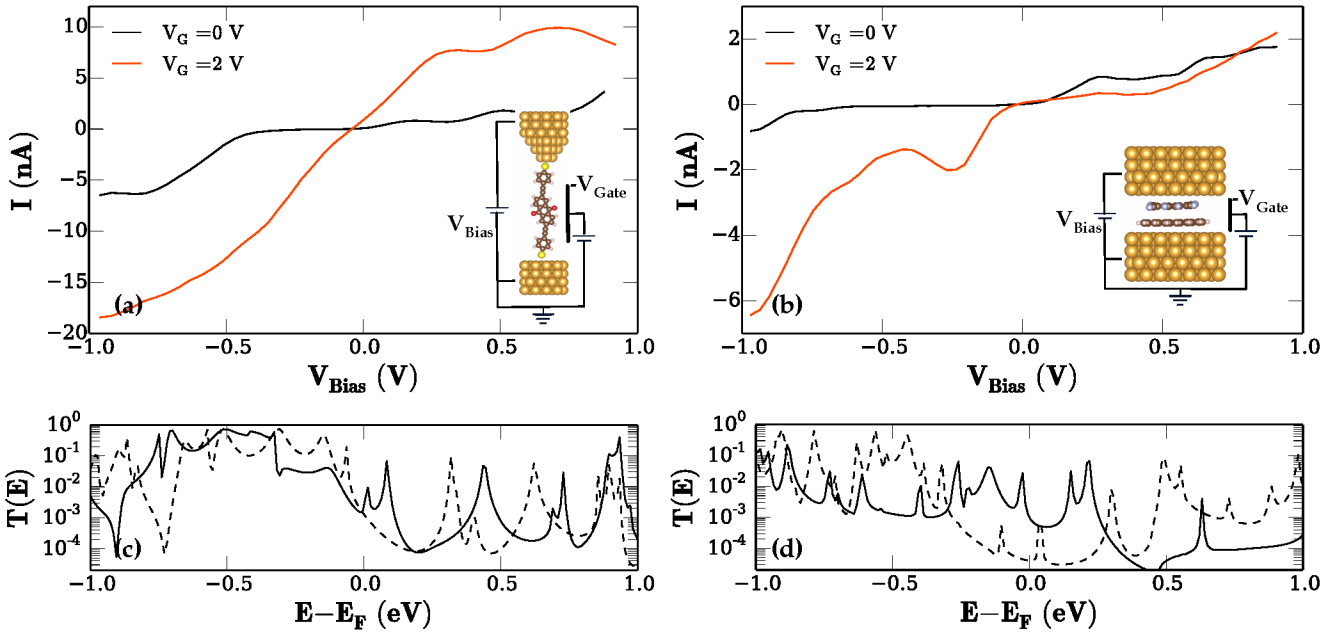

Fig. 3 Calculated transport characteristics of the anthraquinone reference system (a) and the picene-F_4TCNQ model device (b). The reference system shows moderate enhancement of the current in both bias directions. In contrast the picene-F_4TCNQ shows a very strong increase only in reverse-bias region while the forward-bias current is nearly not affected by the external gate field. (c), (d) Respective transmission spectra $T(E)$ for the molecular junctions at zero (solid line) and -0.75 V (dashed line) bias.

an $V_{bias} < -0.8$ V. For anthraquinone junction (Fig. 3a) the current rises more or less symmetrical in case of forward and reverse bias.

This asymmetric behavior gives rise to large current rectification for the picene-F_4TCNQ system. The rectification ratio can be estimated by $R(V) = |I(V)/I(-V)|$[6]. We show a comparison of R for the two systems in the voltage range of interest in Fig. 4b. The highest R are achievable for very low values of V however for real applications voltages between $0.2 - 0.8$ V seem to be more manageable whereas higher bias voltages may lead to rapid degradation in the organic material[18]. From our calculation we obtain rectification rations for $R_{Picene} \approx 20$ which is much higher than the achievable maximum values for R_{AQ}. Moreover for the anthraquinone system we find a steep decrease of the rectification up to ≈ 0.6 V whereas the picene-F_4TCNQ systems shows only a weak dependence in the considered bias range.

For further analysis of the rectifying mechanism we calculated the charge transfer of the picene-F_4TCNQ dimer as a function of the applied bias voltage (see ESI S2). The amount of transferred charge between the molecular subunits clearly depends on the applied bias voltage. Therefore one possibility to rationalize the working principle of the picene/F4TCNQ system as molecular diode is to see the system as a natural pn-junction due to the charge transfer creating anode and cathode side of the molecular stack. The weak rectifying characteristics of the anthraquinone system on the other hand mainly originates from small differences in the coupling of the molecule to the electrodes in combination with moderate evolution of transmission features under applied bias. It is also important to note that compared to the original model for a molecular diode proposed by Aviram and Ratner[1], we observe reversed rectification direction. However this is in qualitative agreement with results reported on different molecular systems[19,20]. In the original model, the rectification mechanism is due to the difference in the tunneling rates between the donor and acceptor part of the molecular structure. In contrast to the DFT-NEGF method used for the present calculations the original model also considered only the HOMO and LUMO molecular orbitals and neglected any bias induced changes of the electronic structure as well as changes in the molecule-electrode coupling. Hence the potential barrier build by the charge transfer inside the picene-F_4TCNQ dimer in combination with the large asymmetry of the molecular transmission features are clearly the reasons for the observed direction and height of the rectification effect.

With its large rectification values the picene-F_4TCNQ - system also outweighs already reported maximum values for other charge transfer and molecular materials by a at least one order of magnitude[4,6]. The low variability of R_{Picene} over a

wide bias range seems more appropriate for real world applications. In consequence we propose the picene-F_4TCNQ system as an molecular material to fabricate organic diodes due to its advantageous forward and reverse bias properties.

Further, a required key element for implementing real molecular circuits is electrical switching. For that reason we compared the response of the two molecular systems to the application of an external gate voltage V_{Gate} as schematically drawn in the insets of Fig. 3. Therefore an additional uniform electric field applied as an external potential was used to model the gate electrode. The I-V curves corresponding to a $V_{Gate} = 2\,\mathrm{V}$ for the respective junctions are also shown as red lines in Fig. 3a and b. Both systems show a rather strong response to the application of an external gate field. For better quantification of the effect the change of the current due to the applied gate voltage $\Delta I = |I_0 - I_{Gate}|$ normalized to current without gate I_0 for both systems is presented in Fig. 4a.

For the anthraquinone junction we find a constant increase of the current for $|V_{bias}| > 0.2\,\mathrm{V}$ between a factor two and three. Other authors report very large on/off ratios for anthraquinone junctions of $> 1 \times 10^3$. However these ratios are achieved by chemical modification of the molecule itself and are potentially irreversible in contrast to the electrical switching presented here. Additionally the already mentioned theoretical [12–15] as well as recently measured data [14] on anthraquinone systems all show almost symmetrical behavior of the I-V characteristics. The additional gate field in the junction induces two main effects. First the gate voltage shifts the molecular energy levels with respect to the energy window in which molecular orbitals contribute to the conduction (see [21]). In the present case the positive gate voltage results in higher number of occupied molecular levels contributing and hence the current increases. The second effect of the gate field is the induction of changes of the electronic structure of the molecule itself. DFT calculations on the anthraquinone molecule with an applied electric field equal to the gate field in the transport calculations indicate that for example the HOMO-LUMO gap of the molecule is reduced and the molecular level alignment changes as well. In Fig. 4a one can see that the amplification of the current due to the gate field for the anthraquinone junction is almost symmetrical with respect to the bias voltage.

The same effects of a gate field also occur in case of the picene junction. However, due to the asymmetric character of the molecular orbitals around the Fermi level the impact on the I-V characteristics is much larger. Under forward bias the current is barely affected by the gate voltage. In fact the current shows even a small decrease. For the reverse bias case however we see a large increase of the current due to gate voltage. We achieve a maximum switching effect by about 5×10^2 for a gate voltage $V_G = 4\,\mathrm{V}$ (see Fig. 4a). In Fig. 4c we additionally show in more detail the dependence of the reverse bias current of the picene-F_4TCNQ junction as a function of the applied gate voltage.

As mentioned before the large energy distance between the LUMO and LUMO+1 leads to almost no transmission for reverse bias. As the gate field shifts the molecular levels we see of course an current increase. However this level shifting alone does not explain the quantity of the observed effect. Here a further mechanism comes into play for the picene-F_4TCNQ system. As discussed in the beginning the picene-F_4TCNQ is a weakly bonded dimer with charge transfer of ≈ 0.2 electrons from the picene to F_4TCNQ. Our DFT calculations on the picene-F_4TCNQ dimer with an electric field applied perpendicular to the stacking direction (= transport direction) reveal that the hybridization of the dimer itself depends strongly on the applied field. The applied electric field allows to tune the hybridization between the dimer components. This allows to lower the LUMO-LUMO+n distance drastically. The result is, that in the reverse bias case with the gate field switched on, the number of levels which account to the conduction is increased. This explains the very large switching ratios of 5×10^2.

In addition we wish to point out, that this behavior corresponds perfectly to the arguments given by Ratner and coworkers [1,2] for achieving molecular rectification. Local weak links in a molecule, given in our case through the hybridization in the picene-F_4TCNQ dimer, can result in large rectification ratios whereas strong bonding suppresses the effect. By direct modification of the hybrid levels using a gate field one can reach effective current switching. We believe that this mechanism is quite general for charge transfer systems and should be applicable to other dimer systems as well [22].

2 Summary

To summarize, the NEGF+DFT studies of the anthraquinone-Au(111) and picene-F_4TCNQ -Au(111) systems have shown that the respective systems exhibit fundamentally different I-V characteristics. The anthraquinone system shows an approximately ohmic behavior for low bias voltages and the application of an external gate field results in an increase of the overall current through the junction which is almost symmetric for positive and negative bias voltages.

Due to hybrid dimer states close to the Fermi level the picene-F_4TCNQ I-V curve is very asymmetric with a pronounced diode-like forward/reverse current behavior. In contrast to the anthraquinone system the effect of an applied gate voltage is about two orders of magnitude larger in the reverse bias than in the forward bias case.

Further, we have shown that the anthraquinone system can also be seen as a electrically controllable switch. However, in terms of achieving maximum switching ratios the picene-F_4TCNQ junction shows a clear benefit and can be seen as an

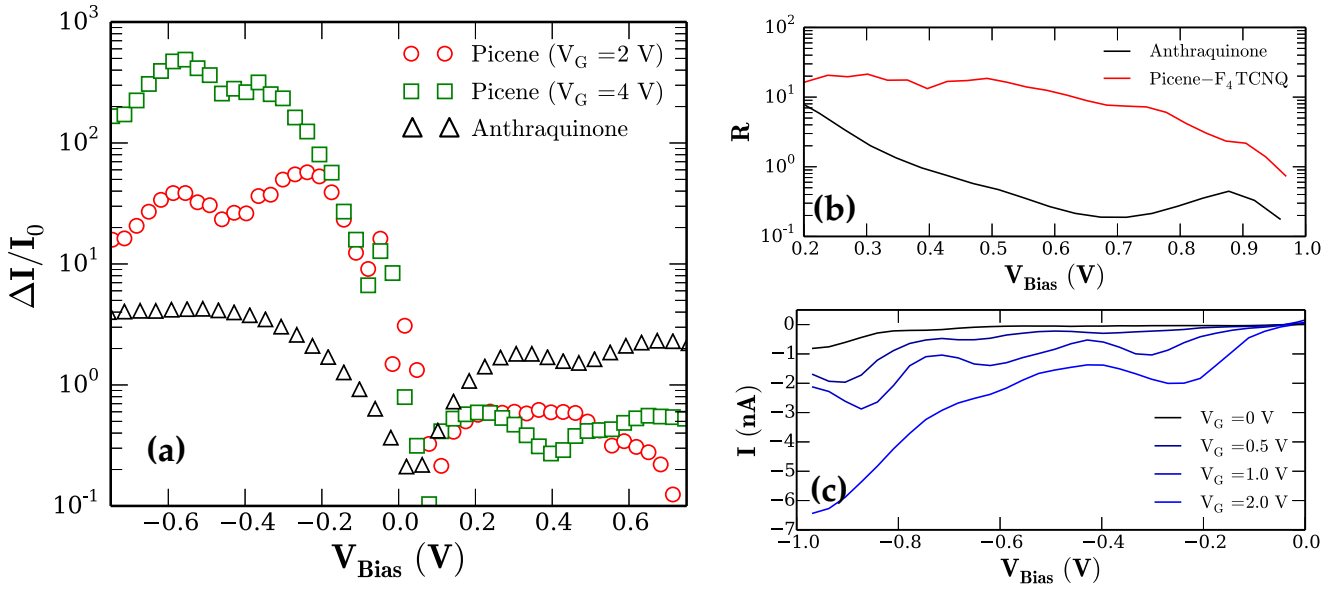

Fig. 4 (a) Current across the picene-F_4TCNQ model junction compared to the anthraquinone reference system as a function of the bias voltage at different gate voltages (I_0 is the respective current without gate voltage). (b) Rectification ratio $R(V) = |I(V)/I(-V)|$ for the two molecular junctions under investigation. (c) Calculated change in the reverse bias current across the picene-F_4TCNQ model junction as a function of the applied gate voltage V_G.

molecular transistor in terms of classical circuit elements. The anthraquinone system on the other hand offers almost symmetric and linear I-V characteristics in the $V_{bias} \pm 0.3$ V range and may be better utilized as operational amplifier.

Consequently, we propose to use the pure organic interface between picene and F_4TCNQ as a straightforward way to manufacture a molecular switch with very large switching ratio or a molecular transistor / amplifier. It should be kept in mind that the presented results rely on the systematic limitations of the applied theoretical model. Especially many-body and strong correlation effects are not included. Nevertheless for the presented cases of low bias voltages the forecasts of the DFT / NEGF method are known to be qualitative correct.

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3 Methods

The ground state electronic structure of the molecules was investigated using the all-electron density functional theory (DFT) NRLMOL program package which achieves a high level of numerical accuracy (see [23,24] and references therein). For the exchange correlation GGA/PBE[25] was used and in all calculations dispersion correction utilizing the DFT-D2 method[26] was included. The geometry of the molecules was optimized using a gradient approach, the relaxation was terminated once all atomic forces were below 0.05 eV/ Å. We applied the nonequilibrium Green's function method (NEGF) for self-consistent calculation of the electron transport properties as implemented in the GPAW code[27,28] to investigate the I-V characteristics of our model devices. For the transport calculations the electronic structure is obtained by DFT calculations using the common approach of constructing a model device where the molecule of interest together with additional electrode atoms (scattering region) are sandwiched between two semi-infinite (metallic) electrodes. In our case we used at least three additional Au(111) layers on each side of the molecule to construct the scattering region followed by a further geometry optimization step of the system where the topmost two gold layers together with the attached molecules were allowed to relax. For the scattering region as well as for the leads a localized double-ζ polarized basis set was used. Schematic

drawings of the used model junctions are shown in the insets in Fig. 3. The whole system can be subject to an external bias and/or gate voltage. The electronic structure of the scattering region and therefore the I-V curves are calculated self-consistently in the presence of such external fields. To support the deductions in this paper we just repeat the key facts of the DFT-NEGF method of use whilst a detailed discussion of the method can be found in the cited literature. The GPAW transport code uses the Green's function of the central region defined by

$$G(E) = [ES - H_C - \Sigma_L(E) - \Sigma_R(E)]^{-1} \quad (1)$$

where S and H_C are the overlap and Hamilton matrix of the scattering region written in the localized basis. $\Sigma_{L/R}$ are the respective self energies of the leads. After self-consistency in the cycle $G \rightarrow n(r) \rightarrow v(r) \rightarrow H_C \rightarrow G$ is reached the transmission function is calculated by

$$T(E,V) = \mathrm{Tr}\left[G(E)\Gamma_L(E)G(E)^{\dagger}\Gamma_R(E)\right] \quad (2)$$

with $\Gamma_{L/R}(E) = i\left(\Sigma_{L/R}(E) - \Sigma_{L/R}(E)^{\dagger}\right)$. Therefore $T(E,V)$ gives the transmission probability of an electron having an energy E under an applied bias (and gate) voltage V. Further the current through the junction is obtained by

$$I(V) = \frac{2e^2}{h}\int_{\mu_L}^{\mu_R} T(E,V)dE \quad (3)$$

were the electronic chemical potentials $\mu_{L/R}$ are connected to the applied bias voltage via $V = (\mu_L - \mu_R)/e$ (e elementary charge)[29]. De facto the current is calculated by integrating the self-consistent transmission function within the bias-dependent energy window spanned by $\mu_{L/R}$.

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