Microscopic Origins of Contact Resistance Reduction by Self Assembled Monolayers

First Principles Computational Study

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Introduction

- Organic devices such as transistors have been limited by contact resistance.
- Self Assembled Monolayer (SAM) at contact interfaces decrease resistance.
- Mechanism of SAM is still ambiguous, and comprehensive models linking physical parameters and device behaviour are unavailable.

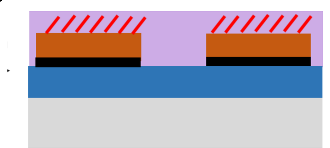

Figure 1: Organic transistor with SAM at contact interfaces

Objectives

1. Identify physical aspects of contact interfaces influencing charge injection
2. Develop a model relating microscopic phenomenon with observed device behaviour

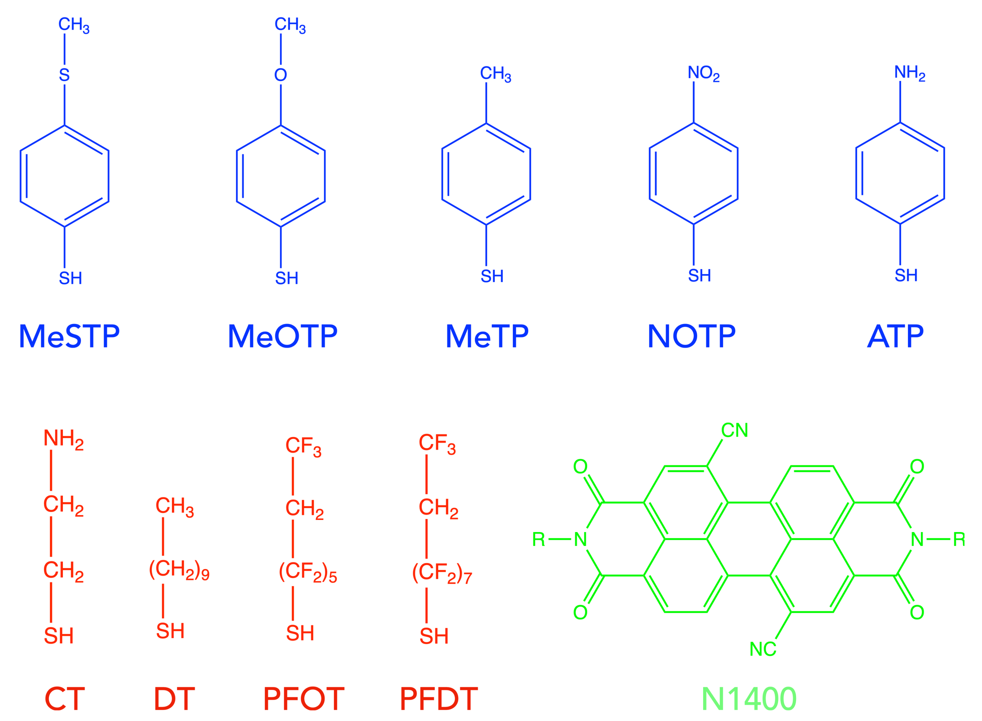

Figure 2: Simulation carried out for various SAM molecules and n-type semiconductor N1400

Orbital Interactions

$$J = \int \alpha_i H \alpha_j dr^3 \quad (1)$$

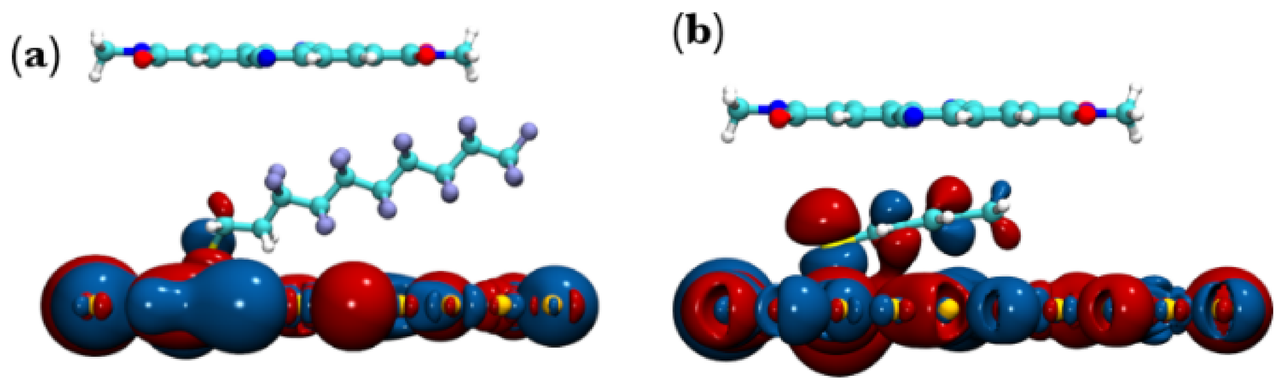

Figure 3: Frontier orbitals of simulated stack with different SAM exhibiting extent of delocalization correlating with device resistance.

Effect of Parameters

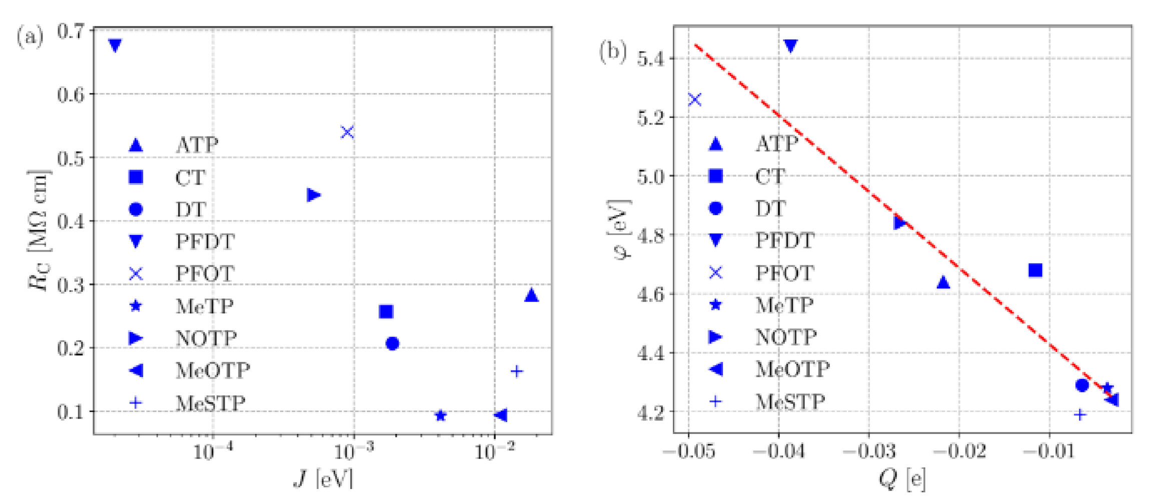

Figure 4: J correlates with resistance for most cases. Q captures the effect of work function.

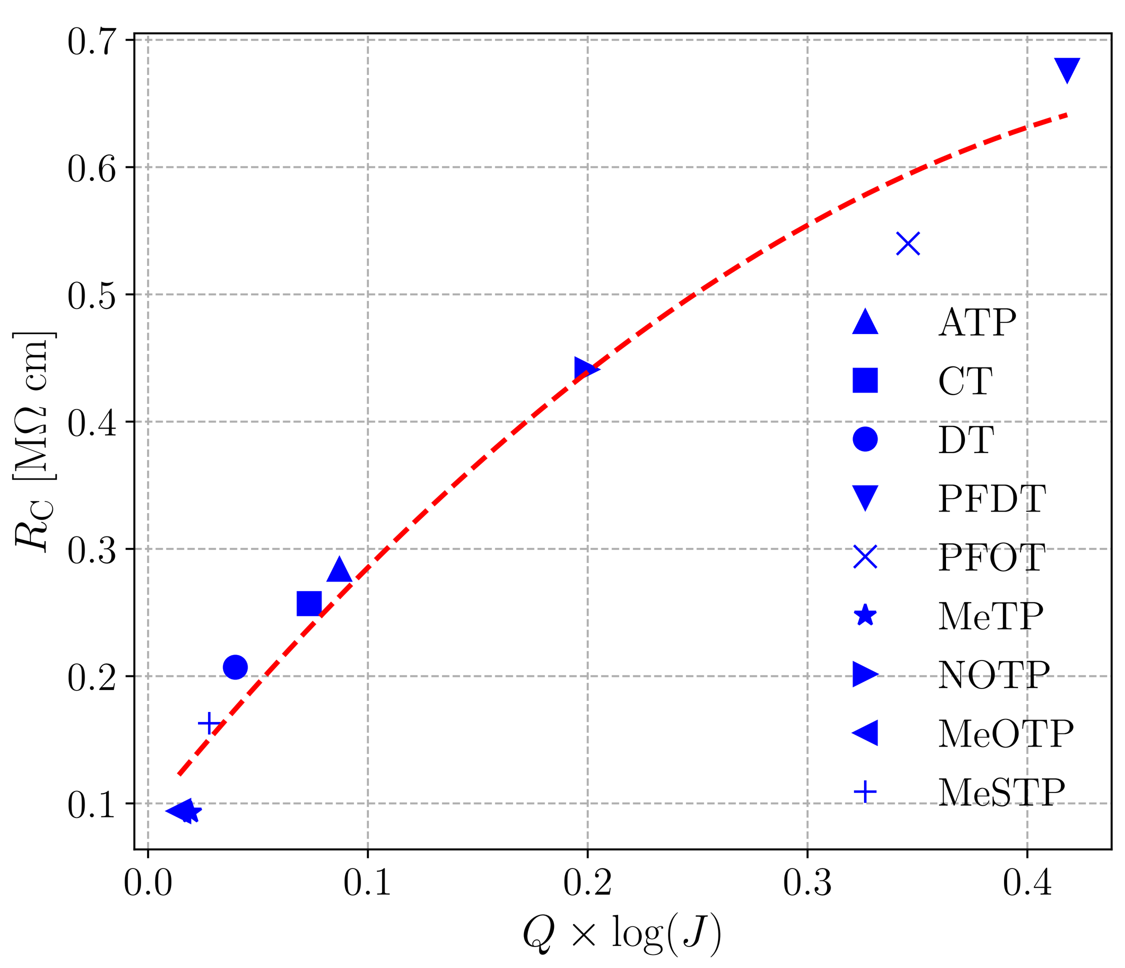

Figure 5: A combination of Q and J efficiently predicts device resistance

Projector Operator Diabatic Method

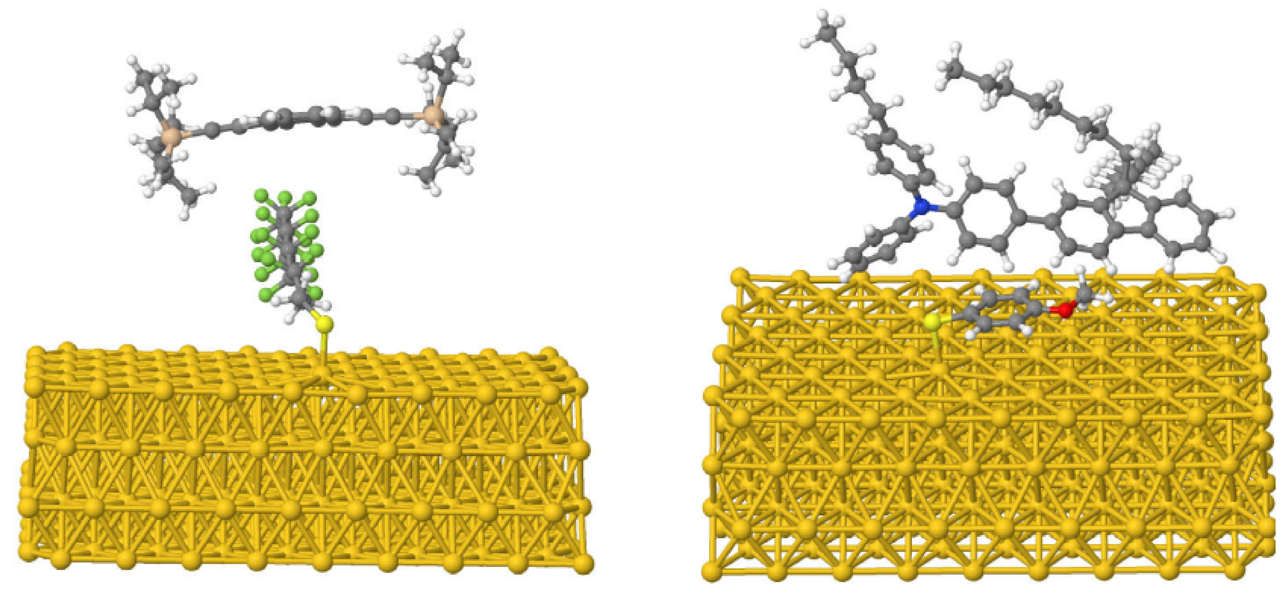

Figure 6: Simulation stack, with p-type OSC

Computations are performed by defining donow and acceptor segments according to POD method. This enables evaluation of separate states and/or orbitals of systems at interface. J is then calculated for for the relavant states and orbitals.

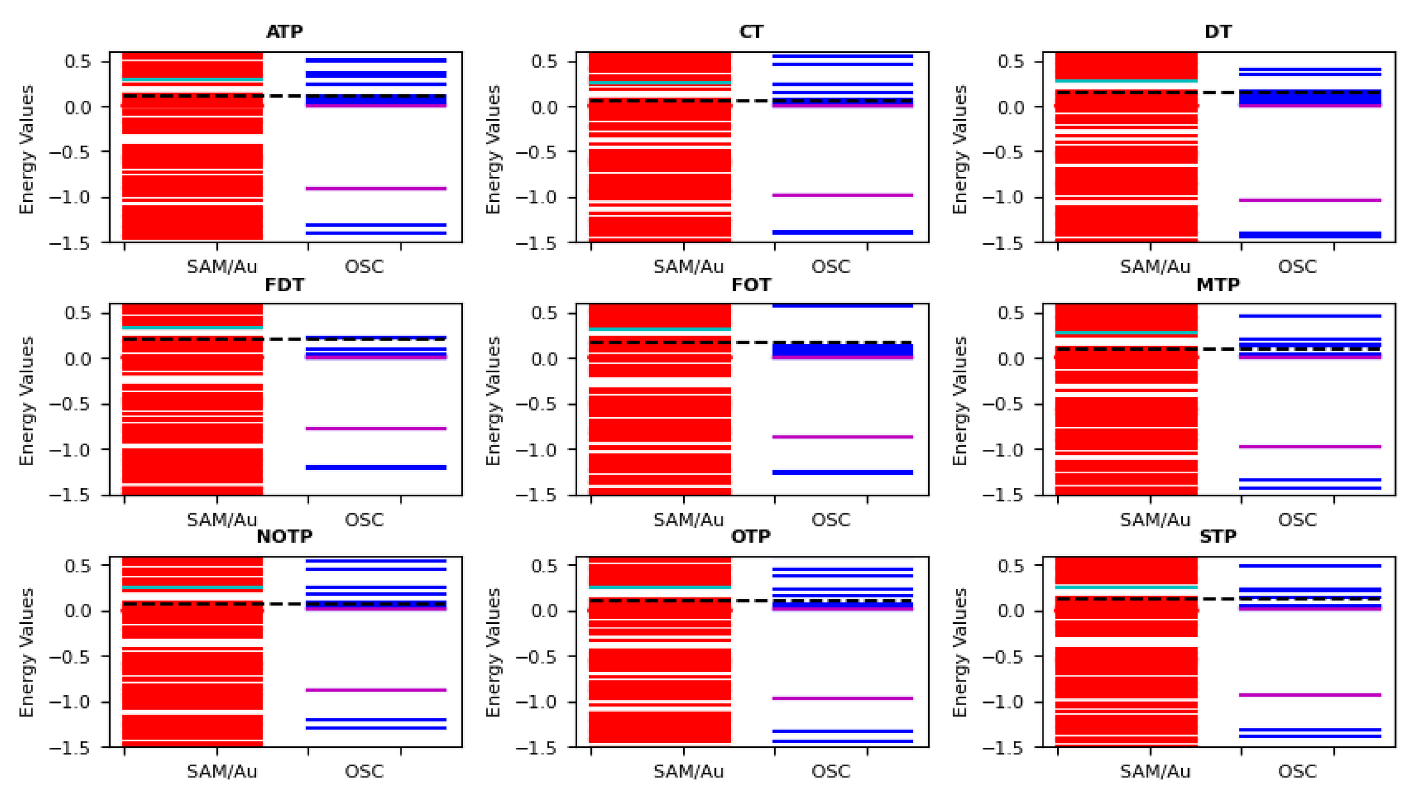

Figure 7: Energy levels for donor and acceptor exhibit the feasibility of charge transfer at interface.

Based ibon energy levels obtained (E), electronic coupling calculated (J), and density of states (n), rate of electron transfer at interface (k_{ET}) are obtained. Marcus-Hush equation gives the rates for electron transfer between molecular and periodic systems.

$$k_{\mathrm{ET}} = \frac{2\pi}{\hbar} \int J^2_{\mathrm{DA}}(E) \frac{1}{1+\exp\frac{E-E_F}{kT}} n(E) \frac{1}{\sqrt{4\pi\lambda kT}} \exp\frac{-(\lambda - \Delta E + q\eta)^2}{4\lambda kT} dE \quad (2)$$

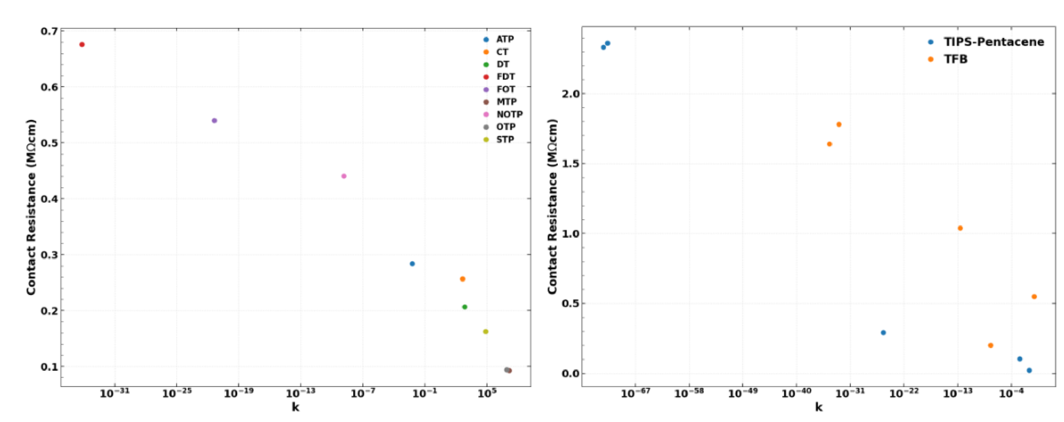

Figure 8: Rates computed for SAM/Au-OSC systems correlate well with measured resistance for N1400. General trends are visible for p-type OSC as well.

Conclusions

- Orbital interactions are crucial to reduction of contact resistance by SAM
- J and Q predict the device resistance of SAM modified devices
- POD enables calculations of energy lvels and consequntly the rates of charge injection
- Rate of charge injection involves orbital interactions as well as energy levels, and forms a comprehensive parameter for prediction of contact resistance.

Other Ongoing Projects

Simulations of charge transfer dynamics of systems including catalysts, polymer-electrode interfaces. Device model based on microscopic phenomenon at OSC-electrode interface.

References

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[2] Kalyani Patrikar, Urvashi Bothra, Valipe Ramgopal Rao, and Dinesh Kabra. Charge carrier doping as mechanism of self-assembled monolayers functionalized electrodes in organic field effect transistors. *Adv. Mater. Interfaces*, 9(1):2101377, 2022.