

First-principles calculation of the effect of adsorbates on field-emission current from single-walled carbon nanotubes

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Although nanotube-based electron emitters have been investigated for over a decade, there are still many unknowns regarding their emission behavior. For example, it has been debated whether they follow the Fowler-Nordheim model. In some cases the observed current saturation has been attributed to the effect of adsorbates on the emitter tip.

The difficulty has been that first-principles modeling of nanotube emitters under realistic experimental scenarios is computationally very expensive. In particular, although many interesting works have been done on the first-principles investigation of the electronic structure of nanotube emitters, much less has been done on transport simulation in these devices and the calculation of emission current. Moreover, most commercial transport solvers for nanoscale devices focus on devices connected between two electrodes and are not capable of treating vacuum (which is part of the structure in the case of an electron emitter) since they use atomic basis sets that do not extend into vacuum.

We present a calculation of the emission current using our newly-developed three-dimensional, real-space transport solver. The advantage of working in the real space is that vacuum can be defined easily. The electronic structure of the nanotube under an applied field is first obtained using a commercial first-principles package (Gaussian 03 [1]) and the obtained Hamiltonian mapped to the real space to form part of the overall device Hamiltonian (which also includes the vacuum section). The current is obtained using a non-equilibrium Greens function method. It is observed that at high fields the current saturates and the current-voltage characteristics indeed do not follow a Fowler-Nordheim trend, even for a pure nanotube with no adsorbates on the tip. Nevertheless, in many real experimental conditions, due to poor vacuum and existence of residual hydrocarbons or other species, adsorbates could play an important role. In particular, hydrogen is one of the most difficult species to remove even at ultra-high vacuum conditions. Here, we explore the emission characteristics of single-walled carbon nanotubes under various coverage levels of hydrogen, nitrogen and some hydrocarbon molecules. It is observed, for example, that covering the surface of the tip with one monolayer

of hydrogen leads to higher turn-on voltages, but significantly raises the highest occupied molecular orbitals and generally increases the emission current by up to about an order of magnitude in the pseudo-exponential part of the current-voltage characteristics.

[1] Gaussian 03, Revision C.02, M. J. Frisch et al, Gaussian, Inc., Wallingford CT, 2004.