## P:33 Computing interfacial properties of polypyrrole on diamond nanoparticles for photovoltaic applications

Petra Matunová [1,2], Vít Jirásek[1], Bohuslav Rezek[1,2]

[1]Institute of Physics of the Czech Academy of Sciences, Czech Republic, [2]Czech Technical University, Czech Republic

Low-cost and high-efficiency renewable energy is nowadays increasing in importance [1]. Diamond-based inorganic-organic hybrid systems may have an immense, yet still mostly unexplored potential in photovoltaic solar cells. That was suggested for instance by previously measured transfer of photo-generated charge between bulk diamond and polypyrrole (PPy) [2–4] or other organic molecules [5]. In this work, we focus on studying interactions of PPy with diamond nanoparticles (so-called nanodiamonds - NDs) by computational methods in order to reveal and better understand effects possibly brought about by the nanoscale features. Density functional theory (DFT) is employed, particularly the B3LYP functional and the 6-31G(d) basis set are used. We compare PPy in chemisorbed and physisorbed configurations on the most probable (111) and (100) ND surface slabs [6,7], having the most common hydrogen- terminated surfaces [8], as well as frequently present oxygen, hydroxyl, carboxyl, and anhydride terminated surfaces. Consistently for hydrogenated and oxidized ND surfaces, calculated binding energies (E_B) indicate exothermic and endothermic character for physisorbed and chemisorbed structures, respectively. For the oxidized ND surfaces, though, the analysis reveals significant role of hydrogen bonds in the physisorption of PPy. In some cases, we obtain more positive values, up to 4.44 eV for physisorbed structures (for PPy on peroxide-terminated 1 × 1 (111) ND surface slab) and up to -0.90 eV for chemisorbed structures (for 1-bond contact of PPy on epoxide-terminated 2 × 1 (111) ND surface slab). Generally, the one-bond contact is energetically more favorable than the two-bond contact. Interaction energies (E_{int}) confirm covalent and non-covalent bonds for all the chemisorbed and physisorbed structures, respectively. Charge transfer (Δq) is observed for all the interfacial structures. The highest charge transfer was obtained for PPy physisorbed on peroxide-terminated 1 × 1 (111) ND surface slab ($\Delta q = 0.23$ e⁻). For the hydrogen-terminated NDs, the charge transfer is the most pronounced in the case of one-bond contact of PPy chemisorbed on 1 × 1 (111) ND surface slab (-0.11 e⁻). Moreover, in a significant number of cases we obtain a spatial separation of HOMO and LUMO at the interface, which seems promising for photovoltaic applications. Simulations addressing an amorphous carbon layer on the surface are beyond the scope of this work. However, they are already in progress in order to model detonation nanodiamond (DND) particles more appropriately. For a better understanding of the observed phenomena and further improvement of the models, simultaneous experimental work is being done, for instance corroborating the bonding character between PPy and NDs [9].

Figure: Schematic of truncated octahedral ND functionalized with various oxygen-containing groups, and PPy chain adsorbed on the surface. (111) ND facets are in green, (100) ND facets are in orange, C atoms of PPy are in gray, H atoms of PPy are in white, and N atoms of PPy are in blue.
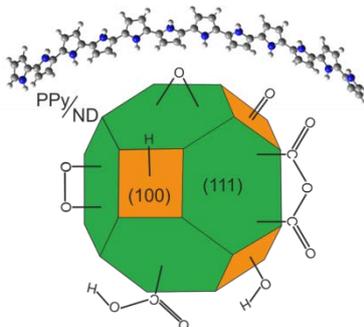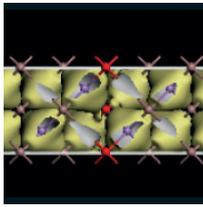
[1]   B. R. Saunders and M. L. Turner, Adv. Colloid Interface Sci. 138, 1 (2008).

[2]   B. Rezek, J. Čermák, A. Kromka, M. Ledinský, and J. Kočka, Diam. Relat. Mater. 18, 249 (2009).

[3]   P. Galář, J. Čermák, P. Malý, A. Kromka, and B. Rezek, J. Appl. Phys. 116, 223103 (2014).

[4]   B. Rezek, J. Čermák, A. Kromka, M. Ledinskỳ, P. Hubík, J. J. Mareš, A. Purkrt, V. Cimrová, A. Fejfar, and J. Kočka, Nanoscale Res. Lett. 6, 1 (2011).

[5]   Y. L. Zhong, A. Midya, Z. Ng, Z.-K. Chen, M. Daenen, M. Nesladek, and K. P. Loh, J. Am. Chem. Soc. 130, 17218 (2008).

[6]   H. Yang, L. Xu, C. Gu, and S. B. Zhang, Appl. Surf. Sci. 253, 4260 (2007).

[7]   D. Petrini and K. Larsson, J. Phys. Chem. C 112, 3018 (2008).

[8]   P. Matunová, V. Jirásek, and B. Rezek, Phys. Status Solidi A 213, 2672 (2016).

[9]   D. Miliaieva, S. Stehlik, P. Stenclova, and B. Rezek, Phys. Status Solidi A 213, 2687 (2016).

P:34 Wigner modelling of surface roughness in quantum wires

P Ellinghaus, J Weinbub, M Nedjalkov, and S Selberherr

TU Wien, Austria

Aggressively scaled More Moore devices, such as FinFETs and nanowire transistors, are designed around the concept of spatial confinement, where electrons are not point-like particles with a continuous spectrum of the momentum: The finite electron size precludes - according to the Heisenberg principle - a well-determined momentum component. Physical processes are usually modelled by a decomposition of the problem into transport and eigenvalue tasks. In quantum wires, the transport is along the wire, where homogeneous conditions are assumed, while the eigenvalue problem in the transverse plane is posed in terms of eigenfunctions and energy subbands. Within this approach shape variations (e.g. roughness) are treated as perturbations which give rise to scattering. Scattering probability models based on the Fermi Golden Rule depend explicitly on the in-plane eigenfunctions, while the subband energies appear in the energy conserving delta function. The eigenvalue problem can be solved either for an ideal wire or with account for the rough interface [1]. A statistical averaging is performed, which gives rise to a roughness-aware, but homogeneous model as long as the probability is independent of the position along the wire. The electron dynamics is captured by the long-time limit of the electron-surface potential interaction process.

In this work, we use the Wigner function approach to simulate the time-dependent electron dynamics in the presence of surface potential variations. The generic process is tunnelling; no artificial borders are introduced. Identical, minimum uncertainty Wigner states [2] $f_w = N \exp\{(r - r_0)^2 / s\sigma^2\} \exp\{(k - k0)^2 2\sigma^2\}$ are periodically injected, with r_0 centered in the source contact of the wire and $\sigma_{x,y} = 2nm$, corresponding to the equilibrium distribution around $k0$ with the effective mass $m* = 0.19$ at $T = 300K$. A signed particle method [3][4] with coherence length $L_x = L_y = 45nm$ is applied. The behavior of currents and densities in the time domain provides rich information about the involved physics. Figure 1 shows an initial penetration in the walls, until the potential modifies the initial distribution with the evolution of the electrons along the channel. The current, calculated with the Ramo-Shockley theorem, linearly increases due to the $5fs$ periodic injection of electron states. After $400fs$, steady-state conditions are reached. Figure 2 compares densities of the ideal and the rough wire. The latter is obtained by superimposing variations of the potential with a correlation function $L_0 \exp\{\Delta x / c_l\}$ [5] on the ideal geometry. The assumption for homogeneous conditions is challenged in both cases, in particular the source and drain regions are well identified in the ideal case. Figure 3 shows the current evolution in the time domain for three different values of $k_{o,y}$. States with a higher $k_{o,y}$ travel faster, therefore a steady-state is reached earlier. Moreover, the