The chemistry and physics of carbon from first-principles, multiscale simulations, and experiments

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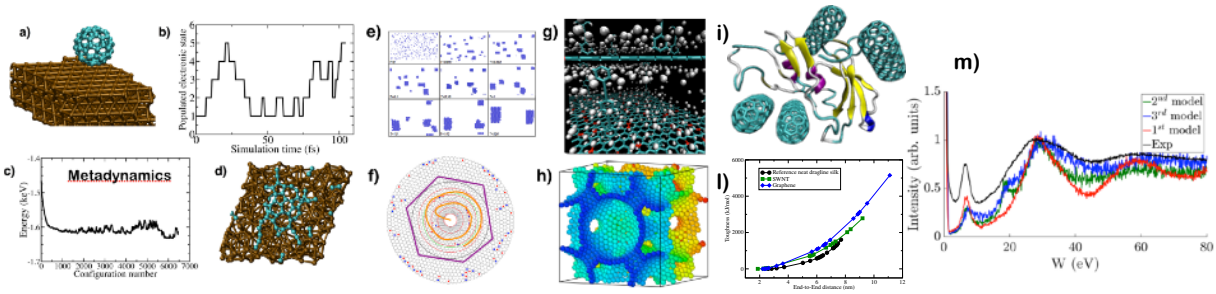
In this talk I will first discuss the processes leading to the room-temperature growth of carbon-based materials, notably silicon carbide (SiC) [1,2] and graphene [3,4], by supersonic molecular beam epitaxy. In particular, I will present both experiments and computational modelling of fullerene impacts on silicon and copper surfaces (Fig. a)) at intermediate-energy regimes (few tens of eV). This collision induces strong chemical-physical perturbations in the system and, for sufficient C_{60} translational energy, disruption of molecular bonds and cage breaking. Characterization of the epitaxial grown materials by a variety of experimental techniques, such as XPS, UPS, Auger, LEED, TEM, and Raman after the collision, demonstrates the potentiality of our approach to grow nanostructured and 2D materials at low temperature. On the theoretical side, we show that in these out-of-equilibrium conditions it is necessary to go beyond the standard implementation of ab-initio molecular dynamics based on the Born-Oppenheimer approximation, which fails to capture the excited-state dynamics. In particular, we analyse the Si- and Cu-C_{60} collision within the non-adiabatic nuclear dynamics framework (Fig. b)), where stochastic hops occur between adiabatic surfaces calculated via time-dependent density functional theory. The theoretical description of C_{60} impacts on metallic and semiconductor substrates will be further analysed by multiscale techniques based on metadynamics (Fig. c), d)) and Kinetic Monte Carlo (Fig. e)).

Furthermore, the discovery of novel energetically stable carbon structures shaped as Beltrami pseudospheres (Fig. f)) for investigating the physics of curved spacetimes [5], the mechanical and gas-sieving properties of pillared graphene oxides [6] and foams [7,8] will be shown (Fig. g), h)). Moreover, we report the observations of silk mechanical properties increment, up to a fracture strength ~5.4 GPa and to a toughness modulus ~1570 J g^{-1}, by incorporating graphene and carbon nanotubes (Figs. i), l)) via spider spinning, after feeding spiders with aqueous dispersions [9].

Finally, Monte Carlo simulations of Reflection Electron Energy Loss spectra (REELS) of diamond and HOP graphite in order to investigate the role of the anisotropic structure in plasmon excitations will be discussed [10]. In our model, elastic scattering between electrons and target atoms is treated via the Mott theory, which is based on the solution of the Dirac equation in a central field. Inelastic interactions between primary beam electrons and the target electron cloud, resulting in bulk excitation and surface plasma oscillations, are described by the dielectric theory developed by Ritchie. In this model, the key quantity for the calculation of the inelastic cross section is the momentum- and energy-dependent dielectric function, which is calculated via a full ab initio approach (Fig. m)).



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