First-principles simulations for nanomaterials

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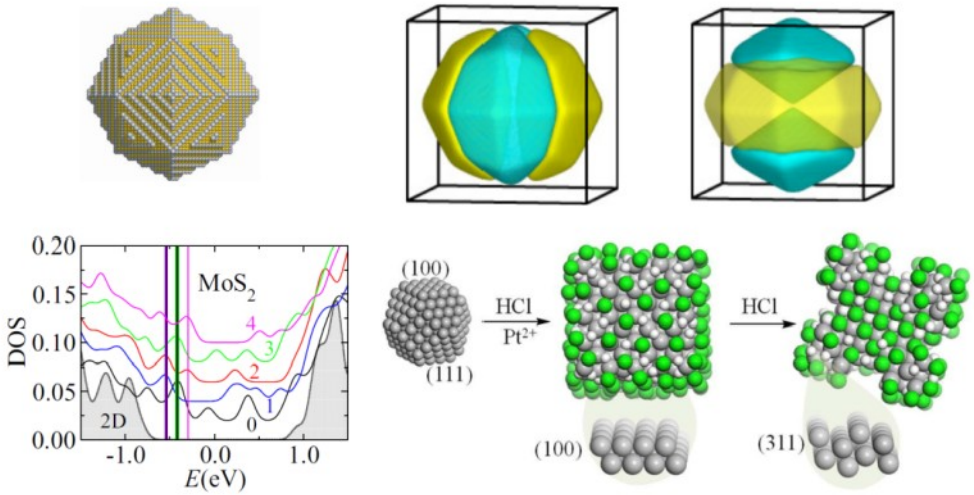
Electronic structure calculations, typically at the level of Density-Functional Theory (DFT), play a key role in the design of new materials, including nanocomposites, heterostructures and nanoparticles. Such complicated structures often demand for multi-scale simulation schemes where DFT couples to classical atomistic or continuous models in order to link the electronic structure to properties of materials. In this talk, I will review this methodology and present results for two classes of hard nanomaterials that dominate current research in materials theory: metal nanoparticles and two-dimensional materials.

Atomistic models of nanoparticles are generated by combining the Wulff construction and surface energies obtained from DFT calculations [3]; these models allow for detailed calculations for any structure-property relationship such as the number of active sites for catalysis [1] or the quantum-confinement levels [4]. This method predicts convex polyhedral shapes for nanoparticles; a recent extension allows for concave polyhedral shapes, too [5]. We studied the well-known problem of metallic edges on two-dimensional semiconductors and found that MoS2 and related materials possess metallic edges. Electrons are localized within 0.5 nm of the zigzag edge, and the energy of these states lies in the middle of the gap of the single-layer [2,6].



<u>From top left, clockwise:</u> *Equilibrium structure for thiol-covered Au nanoparticles [1]; Wavefucntions of the doubly-degerate second excited electronic state in this Au nanoparticle [4]; Shape evolution of Pt nanoparticles in HCl solutions: truncated octahedron to cube to octapod [5]; Electronic Density of states for 2D MoS2 and four different edge terminations [6] showing metallic states in the gap.*

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2. D. Davelou, G. Kopidakis, G. Kioseoglou, *I. N. Remediakis, MoS2 nanostructures: Semiconductors with metallic edges,* Solid State Commun. 192, 42-46, (2014).

3. G. D. Barmparis, Z. Lodziana, N. Lopez and I. N. Remediakis, *Nanoparticle shapes by using Wulff constructions and first-principles calculations,* Beilstein J. of Nanotechn. 6, 361, (2015).

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5. Q. Li, M. Rellán-Piñeiro, N. Almora-Barrios, M. Garcia-Ratés, I.N Remediakis, N. Lopez, *Shape control in concave metal nanoparticles by etching,* Nanoscale 9 (35), 13089-13094 (2017).

6. D. Davelou, G. Kopidakis, E. Kaxiras, I. N. Remediakis, Nanoribbon edges of transition-metal dichalcogenides: Stability and electronic properties, Phys. Rev. B 96, 16, 165436, (2017).