Computational Exploration of Nanoscale Materials with Density Functional Theory: Uncovering Structural, Mechanical, and Electronic Properties

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Density Functional Theory (DFT) has become an indispensable tool for investigating the electronic structure of materials and for exploring their properties at the nanoscale. In this presentation, we first provide an overview of DFT and commonly used approximations within its framework. We then introduce the inverse Kohn-Sham (KS) problem and discuss its usefulness in the development of accurate exchange-correlation functionals. An efficient inversion scheme is presented [1], in which the screening density, linked to the optimal KS potential through Poisson's equation, serves as the variational quantity, enabling access to high-quality KS potentials from accurate electronic densities.

We then highlight a selection of our recent results on materials of contemporary scientific and technological interest:

(a) **Carbon-doped cuprous oxide (Cu_2O):** We analyze the influence of carbon doping on the semiconductor's electronic properties and show that carbon substitution of Cu atoms promotes n-type conductivity.

(b) **Halide, Zr based, "defect" perovskites (A_2ZrX_6) [2,3]:** We assess how composition and doping affect structural stability and the band gap. We considered A = $(CH_3)_3S^+$, $CH(NH_2)_2^+$, NH_4^+, $CH_3NH_3^+$, $(CH_3)_2NH_2^+$, $(CH_3)_3NH^+$, PH_4^+; X = Cl, Br, I.

(c) **Graphene-based, nanoporous membranes for gas separation [4]:** Using transition-state calculations combined with the kinetic theory of gases, we evaluate the performance of single-layer porous-graphene membranes, identifying pyridinic pores of ~0.5 Å diameter as highly promising for selective gas separation.

(d) **Porous carbon-nitride fullerenes [5]:** We examine the structural, vibrational, and thermodynamic stability of carbon–nitride cage molecules that we introduced and summarize their electronic characteristics.

(e) **Interatomic potentials for 2D boron nitride [6]:** We present accurate but computationally efficient, DFT-parameterized interatomic potentials designed for large-scale atomistic simulations.

References:

[1] Bousiadi, S; Gidopoulos, N.I.; Lathiotakis, N. N.; *Phys. Chem. Chem. Phys.* **2022**, *24*, 19279.
[2] Kolokytha, C; Lathiotakis, N.N.; Kaltzoglou, A.; Petsalakis, I. D.; Tzeli, D; *Materials* **2025**, *18*, 726.
[3] Kolokytha, C; Tzeli, D; Lathiotakis, N. N.; *Materials* **2025**, *18*, 3976.
[4] Fthenakis, Z.G.; Petsalakis, I.D.; Lathiotakis, N. N.; *J. Membrane Science* **2025**, *713*, 123339.
[5] Fthenakis, Z.G.; Lathiotakis, N.N.; *Nanoscale Horizons* **2025**, *10*, 1184.
[6] Sgouros, A.P.; Araphatzis, M.; Lathiotakis, N.N.; Papagelis, K.; Kalosakas, G.; *Phys. Rev. B* **2025**, *111*, 155443.