Master 2 project proposal

Analyzing metastable growth pathways of CuPtNi nanoalloys by coupling molecular dynamics, X-ray scattering and deep learning

Host laboratory	ICMN, UMR 7374, CNRS – Université d'Orléans
Supervision	Daniel Förster (atomistic simulation and machine learning), MCF; Claudia de Melo Sanchez (MBE growth and synchrotron characterization), MCF
Duration	6 months

Platinum-based nanoalloys (PtNi, PtCu, CuPtNi) are reference catalysts, in particular for the oxygen reduction reaction. In the CuPtNi system, partial substitution of platinum by copper and nickel lowers cost while tuning activity through strain and d-band position. Functional properties are governed by coupled parameters: size and morphology (truncated octahedra, icosahedra, decahedra), composition, and chemical order (disordered solid solution, core–shell structures, $L1_0$ or $L1_2$ phases, surface segregation).

Grown out of equilibrium by sequential deposition or co-deposition in molecular beam epitaxy (MBE), the particles follow metastable pathways set by the competition between deposition kinetics, surface diffusion and intra-particle atomic diffusion. The resulting states (composition gradients, partial order) are generally not those of the equilibrium phase diagram, and remain poorly understood. Synchrotron X-ray scattering can follow them in situ, but the measured intensity is an average over a heterogeneous, correlated population. Extracting the underlying distributions is an ill-posed inverse problem that classical methods, which assume a homogeneous population or independent distributions, treat only approximately.

The internship proposes to solve this problem through a complete simulation → scattering → machine-learning approach:

(i) Build, by molecular dynamics with a TBSMA potential already parametrized for Cu, Pt, Ni and the cross interactions, a database of 1–4 nm particles spanning the ternary diagram: equilibrium morphologies and chemical configurations (solid solution, Pt@CuNi and Cu@PtNi core–shell, ordered phases, surface segregation), together with atom-by-atom growth runs that reproduce the experimental protocols (Cu on PtNi seeds, Pt on CuNi seeds, co-deposition) at different fluxes and temperatures.

(ii) Compute individual $I(q)$ profiles with the Debye equation,

$$I(q) = \sum_i \sum_j f_i(q)\, f_j(q)\, \frac{\sin(q r_{ij})}{q r_{ij}},$$

averaging over thermal configurations, on the q grids of the synchrotron beamlines used.

(iii) Describe realistic populations by a joint distribution $P(D, x, \chi \mid \theta)$ of size, composition and order state, form aggregated spectra $I_{\mathrm{agg}}(q;\theta) = \sum_i w_i(\theta)\, I_i(q)$, and sample θ over physically reasonable ranges (with counting noise and background as data augmentation) to build the training set.

(iv) Train a 1D convolutional network that maps $\log I(q)$ to the parameters of θ, then apply it to already acquired (GI)WAXS/(GI)SAXS time series for the three MBE protocols, in order to reconstruct the evolution of mean size, composition heterogeneity and core–shell or ordered fractions during growth.

The molecular-dynamics and Debye codes as well as the experimental data are available in the laboratory. The closed experiment–simulation loop will identify the metastable states taken by each system and the conditions (temperature, composition, deposition sequence) that stabilize them.