Theory of electrochemically driven metal nucleation on diamond electrodes

PhD Opening, Diamond Science Technology Centre for Doctoral Training (Diamond CDT)

Supervisor: Dr. Reinhard J. Maurer, Department of Chemistry and Centre for Scientific Computing, University of Warwick
Collaborator: Prof. Julie V. MacPherson, Department of Chemistry, University of Warwick

Background: Electrochemical nanoparticle deposition is an important method to grow thin films and nanoarchitectures for applications in electronics, heterogeneous catalysis, nanomagnetism, and quantum computing. However, especially the initial stages of electrochemical nucleation and the underlying mechanisms of particle growth are not well understood. Recent experimental evidence suggests that the electrochemical potential drives a diverse range of elementary processes that involve adsorption, on-surface and on-cluster diffusion, and staged atom and cluster amorphisation and recrystallisation. The existence of these processes either is missing from or directly contradicts established classical and atomistic theories of nanoparticle growth and nucleation. [1] This project is part of a collaborative experiment-theory effort to develop modern atomistic theories of atom-by-atom particle growth, which will guide the future controlled design of catalytically-active nanostructures.

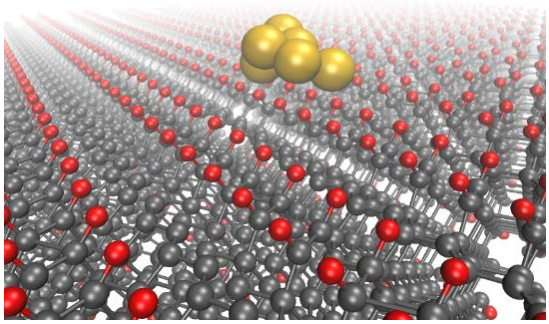

Figure 1 Au_6 nanoparticle adsorbed on an oxidized Diamond (110) surface.

Methodology and work programme: The successful applicant, will use ab-initio simulation based on Density Functional Theory [2] and the highly-efficient Density-Functional Tight-Binding method [3] to study the initial stages of nanoparticle growth during electrodeposition and the stability of nanoparticles of different sizes on oxygen-passivated diamond (see Figure 1). The candidate will apply a range of computational methods including global optimization, ab-initio molecular dynamics, ab-initio thermodynamics, and electrochemical simulations at constant potential [4,5] to study structural, thermodynamic, and kinetic aspects of electro-nucleation on diamond. This will include:

- Constructing a model for oxygen-passivated diamond under varying electrochemical bias.
- Systematic construction of a database of elementary reactions involved during electrochemical nanocluster growth
- Analysis of the reaction dynamics during electrochemically-driven nanoparticle formation
- Construction of a kinetic rate models to predict the critical nucleus, average particle shape and size as a function of applied electrochemical potential

This project will involve proficient use of standardized electronic structure theory software, computations on national and international-scale high-performance computing facilities, and enhanced data analysis and visualization practices. The candidate will be in regular exchange with experimental collaborators, which will provide a two-way feedback to guide experimental and theoretical progress on this project.

References:

[1] H. E. M. Hussein, R. J. Maurer, H. Amari, J. J. P. Peters, L. Meng, R. Beanland, M. E. Newton, J. V. Macpherson, manuscript in preparation (2018)
[2] V. Blum, V. *et al., Comp. Phys. Commun.* **180,** 2175–2196 (2009).
[3] M. Stöhr, G. S. Michelitsch, J. C. Tully, K. Reuter, and R. J. Maurer, *J. Chem. Phys.* **144**, 151101 (2016)
[4] K. Chan and J. K. Norskov, *J. Phys. Chem. Lett*. **6**, 2663-2668 (2015)
[5] S. Sakong, A. Groß, *ACS Catalysis* **6**, 5575-5586 (2016)

Interested candidates should contact Dr. Reinhard Maurer (r.maurer@warwick.ac.uk).
Further information can be found on the Diamond CDT webpage (https://warwick.ac.uk/fac/sci/dst/).