An *ab initio* journey towards the molecular-level understanding of subnanometric metal clusters

From He nanodroplet-mediated surface deposition to the interaction with sunlight

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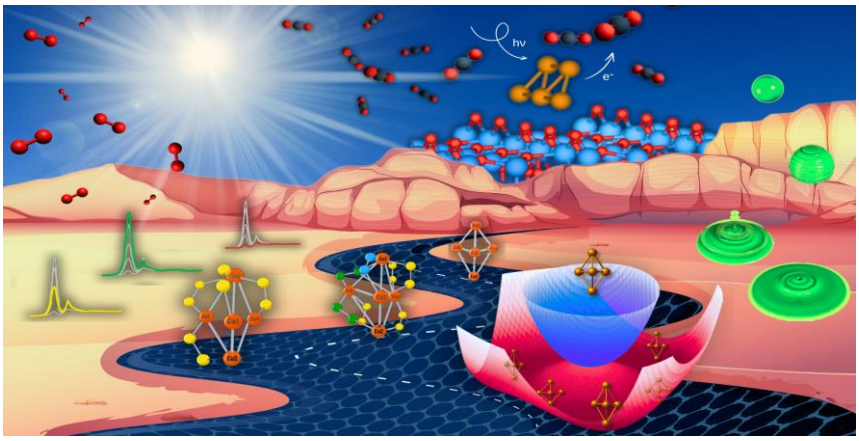
Current advances in synthesizing and characterizing atomically precise monodisperse metal clusters (AMCs) at the subnanometer (Angstrom) scale have opened fascinating possibilities in new quantum materials research. Their quantized 'molecule-like' electronic structure showcases unique stability and physical and chemical properties different from those of nanoparticles and bulk materials. The COST Action CA21101 "COSY" leverage a strategic position in the synthesis of atomically precise metal clusters in various environments (gas-phase, solution, within helium nanodroplets, and in biologically relevant media), their characterization via. e.g., transmission electron microscopy and state-of-the-art computational methods (*ab initio*, DFT and molecular dynamics). When integrated into materials that interact with environmental molecules and sunlight, AMCs may exhibit enhanced (photo)catalytic activity, electronic properties, and even optical behavior. Their tiny size (below 1 nm) makes free AMCs amenable to atomic-scale modelling using at a high level of *ab initio* theory, even including nonadiabatic (e.g., Jahn-Teller) effects. Surfaces supported AMCs can routinely be modelled, enabling real-time molecular dynamics simulations on the picoseconds scale and the study of their optical properties. All these experimental and computational-theoretical efforts aim to achieve a molecular-level understanding of the stability and properties of AMCs as function of their chemical composition, size, and structural fluxionality in different thermodynamical conditions (temperature and pressure). In this talk, covering the topics af COST Action "COSY" Groups, the potential of state-of-the-art *ab initio* modelling will be emphasized through an illustrative overview of recent studies of free and surface-supported AMCs, starting with the helium nanodroplet-mediated surface deposition of single metal atoms, going through the enhancement of the optical properties of titanium dioxide surfaces, and ending with a polarization phenomena in supported silver cluster-induced surface polarons.



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