

Towards the 'Superatom': Experiments with Clusters in the Gas Phase, in Liquids and on Surfaces

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Metal clusters show a great sensitivity to electronic shell closure and geometric symmetry effects. Each constituting atom contributes valence electrons to the entire cluster; shell-closure depends on the stoichiometry of the metal atoms and thus on the clusters chemical composition. Doping, in fact, represents a powerful means to tune cluster properties to specific applications [1]. By designing clusters of the right size and right chemical composition it should be possible to create 'superatoms' of increased chemical stability that could, for instance, replace noble metal atoms in catalysts. However, the locations of the dopant atoms are also important because a foreign atom put in the right place can minimise strain and thus lower the total energy or provide a binding link to a surface.

Consequently, a major requirement for experiments is to provide a method of synthesis with control over (i) the stoichiometry and (ii) the geometrical location of the constituents. In this presentation I will introduce the method of cluster-aggregation to produce 'superatoms' and discuss examples of possible experiments. The method uses liquid helium (or other liquid-like) droplets which pass through plumes of metal vapours and sequentially pick-up single metal atoms which become trapped inside the droplets and agglomerate into clusters. When the droplets then pass through a vapour of a different metal these atoms will land on the surface of the previously built cluster thus providing radial control. Reversing the pick-up order will lead to clusters which are doped in the centre. A further benefit of the method of cluster aggregation is that it allows us to soft-land the embedded clusters on a surface because the kinetic energy will be completely absorbed by the host droplet leaving the bare metal cluster behind. Finally, the method allows us to cage clusters by a protective shell to prevent, for instance, fragmentation and the structure of such core-shell clusters can be assessed spectroscopically. In a proof of principle experiment we could show that single oxygen molecules were caged by two argon layers within a neon cluster [2]. This was possible, because each layer showed a specific fluorescence band and even the interface layer showed a distinct spectroscopic fingerprint.

I will discuss the advantages of our approach to progressively investigate cluster-host interactions on the example of PdAg_n clusters. Recent calculations have identified clusters bound to colour center defects on MgO (100) with magic-like properties [3]. These clusters represent promising candidates to achieve luminescence or functionality. The generation of these clusters by aggregation inside helium droplets will make it possible to investigate their luminescence in the gas phase as well as to land them on defective MgO surfaces and investigate their electronic properties by STS. Furthermore, I will introduce luminescent Si nanoparticles in aqueous suspension that were created by our method [4] as well as Fe nanocrystals landed on carbon surfaces.

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