

# Soft landing and particle coverage key to keeping or losing charge on surfaces

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(Phys.org)—Producing sustainable energy demands materials with specific physical and chemical properties that are controlled by the size and electrical charge of small metal particles, and scientists at Pacific Northwest National Laboratory have discovered how to precisely control both properties. Solid catalysts are typically made of tiny metal clusters dispersed on a supporting material. To more effectively distribute the clusters on the support, a layer of "stringy" molecules may be used to tether the clusters to the surface. The PNNL team found that the properties of these strings control the number of charges carried by the catalytic clusters dispersed on top of the layer.

"When you are developing catalysts, charge matters," said Dr. Julia Laskin, a physical chemist at PNNL who led this research. "We need better tools to measure charge states of catalytic molecules on surfaces."

Catalysts are intricate materials designed to increase the efficiency and reduce the waste associated with creating products such as fuels, plastics and pharmaceuticals. Improving catalysts can allow fuels and other materials to be made with less energy and less waste. Current solid catalysts that act on liquid or gaseous reagents are often composed of poorly defined metal nanoparticles dispersed randomly on supporting materials with large surface areas. In contrast, the next generation of catalysts will be formulated from controlled mesoscale assemblies of [metal clusters](#) with properties that depend on the number of [metal atoms](#) and charge state. To assemble well-defined metal clusters into mesoscale structures for catalysis, techniques are needed that provide precise

control over the size, charge state, and density of metal clusters on surfaces.

"Our approach provides atom-by-atom control of the size and electron-by-electron control of the charge state of metal clusters on surfaces," said Dr. Grant Johnson, a physical chemist involved in the study and former Linus Pauling Fellow who recently joined the Laboratory as a full-time scientist.

When designing catalysts or other energy-related materials, a cook and look approach is often used. Scientists take likely materials and combinations, apply expertise and intuition, and synthesize the new materials that are needed. This process is expensive and time consuming. Fundamental knowledge and atomically [precise control](#) are needed to rationally design the materials scientists want. Enter the PNNL team.

The research comes in two parts: synthesis of the metal clusters and soft landing of size-selected clusters onto surfaces. The clusters used in the study are triply charged positive ions (+3) less than 1 nanometer in diameter. They contain exactly 11 gold atoms.

"The greatest challenge wasn't the synthesis, but the characterization - finding out how many charges the clusters retain after they are soft-landed on surfaces," said Thomas Priest, an undergraduate intern who worked on the study. "It's quite exciting to observe how different they are depending on the properties of the surface." Priest, who was second author on two journal articles describing research done during his internship, synthesized the solutions of [gold clusters](#). Priest is now working on his master's degree in engineering at the University of Louisville.

The surfaces onto which the gold clusters were deposited are a base of gold with a monolayer of molecules containing a hydrocarbon chain and

terminated with different functional group, such as  $-\text{CH}_3$ ,  $-\text{CF}_3$  or  $-\text{COOH}$ . By varying the terminal functional group, the polarity of the molecules was tuned from little to extreme. The polarity of the molecules determines the propensity of the monolayer to tunnel electrons at a certain potential.

Electron tunneling through the layer has a strong effect on the charge of soft-landed multiply charged [cluster](#) ions on the surface. For example, when a mildly polar hydrocarbon-based monolayer is used, the soft landed 3+ gold clusters gain up to three electrons from the surface, thereby becoming neutral. In contrast, on the highly polar fluorine-based surface, the gold clusters keep their 3+ charge, gaining no electrons from the surface. The polar molecules in the monolayer introduce an interface dipole, essentially a charge barrier between the surface and the cluster.

Following this discovery, the researchers went on to see if covering the surface in a denser layer of multiply charged gold clusters changed the clusters' charges. From a charge standpoint, the whole result suddenly changed. On the fluorine-based surface in the earlier experiments, all of the clusters retained their +3 charge. Now, the surface had a center of gold clusters with a +1 charge and, at the edges, the clusters were predominately +2 and +3.

Why did this happen?

"The greater number of multiply charged clusters deposited on the surface built up a sufficient potential to allow the electrons from the [surface](#) to tunnel to the gold clusters, thereby reducing their charge state," explained Johnson. "Understanding how these tunneling barriers break down gives scientists the knowledge they need to control material's charges."

Laskin and Johnson are now using this knowledge to prepare specialized

mesoscale arrays of well-defined metal clusters on surfaces. These arrays could have applications in fuel cells, and efficient energy sources.

**More information:** Johnson, G. et al., Charge Retention by Gold Clusters on Surfaces Prepared Using Soft Landing of Mass Selected Ions. *ACS Nano*, 2012, 6(1):573-582. [DOI: 10.1021/mn2039565](https://doi.org/10.1021/mn2039565)

Johnson, G. et al., Coverage Dependent Charge Reduction of Cationic Gold Clusters on Surfaces Prepared Using Soft Landing of Mass-selected Ions. *The Journal of Physical Chemistry C*, 2012, 116(47):24977-24986. [DOI:10.1021/jp308795r](https://doi.org/10.1021/jp308795r).

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