When aluminum outshines gold: Research details aluminum's valuable plasmonic properties

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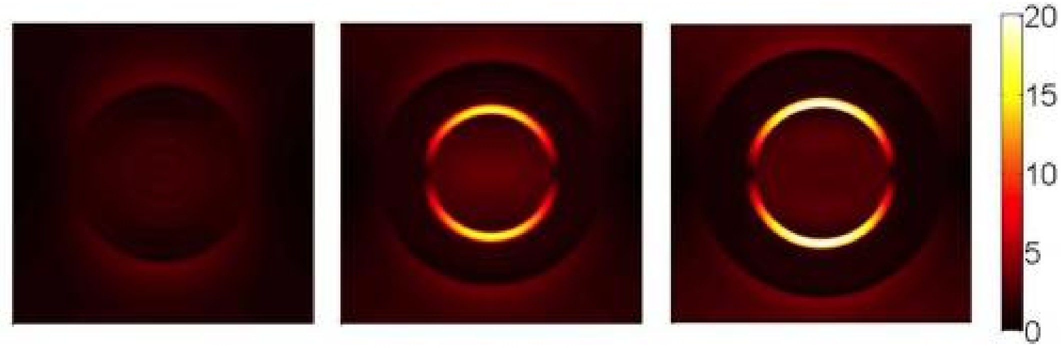

Induced electric fields in Rice University computer models of aluminum nanomatryushkas show that at low gap distances (left), the charge transfer between the core and shell are so large, the system essentially behaves as a solid sphere. At higher gap distances (center and right), the core and shell show stronger individual plasmonic effects. The scale bar shows the strength of the induced field divided by the incoming field strength. Credit: Vikram Kulkarni/Rice University

(Phys.org) —Humble aluminum's plasmonic properties may make it far more valuable than gold and silver for certain applications, according to new research by Rice University scientists.

Because aluminum, as nanoparticles or nanostructures, displays optical resonances across a much broader region of the spectrum than either gold or silver, it may be a good candidate for harvesting solar energy and for other large-area optical devices and materials that would be too expensive to produce with noble or coinage metals.

Until recently, aluminum had not yet been seen as useful for plasmonic applications for several reasons: It naturally oxidizes, and some studies have shown dramatic discrepancies between the resonant "color" of fabricated nanostructured aluminum and theoretical predictions.

The combined work of two Rice labs has addressed each of those hurdles in a pair of new publications.

One paper by the labs of Rice scientists Naomi Halas and Peter Nordlander, "Aluminum for Plasmonics," demonstrates that the color of aluminum nanoparticles depends not only on their size and shape, but also critically on their oxide content. They have shown that, in fact, the color of an aluminum nanoparticle provides direct evidence of the amount of oxidation of the aluminum material itself. The paper appears in the American Chemical Society (ACS) journal *ACS Nano*.

Manufacturing pure aluminum nanoparticles has been a roadblock in their development for plasmonics, but the Halas lab created a range of disk-shaped particles from 70 to 180 nanometers in diameter to test their properties. The researchers found that while gold nanoparticles' plasmons resonate in visible wavelengths from 550 to 700 nanometers and silver from 350 to 700, aluminum can reach into the ultraviolet, to about 200 nanometers.

When an electromagnetic wave (left) hits a nanomatryushka (center and right) -- a solid core inside a hollow shell -- the size of the gap determines the strength of the plasmonic response. If the gap is sufficiently small, quantum tunneling through the gap allows plasmons to resonate as though the core and shell are a single particle, dramatically changing their response. Credit: Vikram Kulkarni/Rice University

The labs also characterized the weakening effect of naturally occurring but self-passivating oxidation on aluminum surfaces. "For iron, rust goes right through," Nordlander said. "But for pure aluminum, the oxide is so hard and impermeable that once you form a three-nanometer sheet of oxide, the process stops." To prove it, the researchers left their disks exposed to the open air for three weeks before testing again and found their response unchanged.

"The reason we use gold and silver in nanoscience is that they don't oxidize. But finally, with aluminum, nature has given us something we can exploit," Nordlander said.

The second paper by Nordlander and his group predicts quantum effects in plasmonic aluminum that are stronger than those in an analogous gold structure when in the form of a nanomatryushka, multilayer nanoparticles named for the famous Russian nesting dolls. Nordlander discovered the quantum mechanical effects in these materials are strongly connected to the size of the gap between the shell and the core. The paper appeared recently in the ACS journal *Nano Letters*.

"In addition to being a cheap and tunable material, it exhibits quantum mechanical effects at larger, more accessible and more precise ranges than gold or silver," Nordlander said. "We see this as a foundational paper."

Nordlander used computer simulations to investigate the discrepancies between classical electromagnetics and quantum mechanics, and precisely where the two theories diverge in both gold and aluminum nanomatryushkas. "Aluminum exhibits much more quantum behavior at a given gap size than gold," he said. "Basically for very small gaps, everything is in the quantum realm (where subatomic forces rule), but as you make the gap larger, the system turns to classical physics."

By small, Nordlander means well below a single nanometer (a billionth of a meter). With the gap between core and shell in a gold nanomatryushka at about half a nanometer, he and lead author Vikram Kulkarni, a Rice graduate student, found electrons gained the capability to tunnel from one layer to another in the nanoparticle. A 50 percent larger gap in aluminum allowed for the same quantum effect. In both cases, quantum tunneling through the gap allowed plasmons to resonate as though the core and shell were a single particle, dramatically enhancing their response.

The calculations should be of great interest to those who use nanoparticles as probes in Raman spectroscopy, where quantum tunneling between particles can dampen electric fields and throw off classical calculations, he said.

Nordlander noted that Kulkarni's algorithm allowed the team to run one of the largest quantum plasmonics calculations ever performed. They used the power of Rice's BlueBioU supercomputer to track a massive number of electrons. "It's easy to keep track of two children, but imagine if you had more than a million," he said.

Lead authors of "Aluminum for Plasmonics" are Rice graduate students Mark Knight and Nicholas King. Co-authors include graduate student Lifei Liu and Henry Everitt, a chief scientist at the U.S. Army's Charles Bowden Research Lab, Redstone Arsenal, Ala., and an adjunct professor at Duke University. The research was supported by the Robert A. Welch Foundation, the National Security Science and Engineering Faculty Fellowship, the Air Force Office of Scientific Research, the National Science Foundation's Major Research Instrumentation Program, the Army's in-house laboratory-independent research program and the Army Research Office.

Rice alumnus Emil Prodan, an assistant professor of physics at Yeshiva University, New York, is co-author of "Quantum Plasmonics: Optical Properties of a Nanomatryushka."

More information: Read the abstract for "Aluminum for Plasmonics" at pubs.acs.org/doi/abs/10.1021/nn405495q
Read the abstract for "Quantum Plasmonics: Optical Properties of a Nanomatryushka" at pubs.acs.org/doi/abs/10.1021/nl402662e

Provided by Rice University

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