



Thinner silver films, stronger nonlinear response

Researchers at the University of Vienna, the University of the Basque Country, and ICFO have presented a novel strategy for enhancing nonlinear optical responses in two studies, one published in *Nature Communications* and the other in *Advanced Materials*. The researchers use atomically thin silver films rather than bulk materials, and pattern these films laterally for an additional enhancement.

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Nonlinear processes are ubiquitous in modern scientific research. We could not generate certain laser frequencies or entangled photon pairs without them, for instance. Scientists have typically employed long bulk materials as the nonlinear media, so that light can interact extensively with them to generate a strong enough nonlinear response. This, however, works against the push toward miniaturized, on-chip photonic devices.

Now, researchers at the University of Vienna and the University of the Basque Country, together with researchers at ICFO, **Dr. Saad Abdullah, Dr. Andrew P. Weber, Dr. Alvaro**

Rodriguez Echarri, Dr. Fadil Iyikanat, and Dr. Vahagn Mkhitarian, led by ICREA Prof. F. Javier Garcia de Abajo, have instead used an atomically thin silver platform to produce a strong nonlinear response.

First, in a Nature Communications article, they **harnessed the internal properties of ultrathin silver films**. The researchers grew ultrathin, single-crystal silver films epitaxially on silicon wafers and shone mid-infrared laser pulses through them, which generated second-order harmonics (a nonlinear process where light at one frequency is converted into light at double that frequency). The team then found that, even though less material generally leads to weaker nonlinear signals, **the thinner the films, the stronger the second-harmonic signal**.

The reason for this counterintuitive behavior lies in the phenomenon of quantum confinement. In bulk metals, the electrons can have any energy within continuous energy bands. But in ultrathin layers, which lie at the boundary between bulk and two-dimensional materials, these energies become quantized, approaching unevenly spaced discrete energy levels similar to those present in an atom. As the film gets thinner, these levels become more separated, and this in turn promotes the nonlinear response and increases the efficiency of second-harmonic generation.

Then, in an Advanced Materials study, the team **patterned these films laterally into periodic arrays of nanoribbons and nanotriangles**. Such nanopatterns make the conduction electrons oscillate collectively in response to the incident light, forming so-called plasmons. For particular combinations of nanostructures and wavelengths of the incoming light, these plasmons are confined to nanoscale volumes, which, as the researchers showed, increases the nonlinear signal by generating stronger second harmonics.

“We have combined two different confinement mechanisms in the same atomically thin silver platform,” says Dr. Saad Abdullah, first author of the article. “In this way, we have shown that significant nonlinear functionality can be obtained from structures only a few nanometres thick, which is attractive for highly compact nonlinear optical devices and future on-chip photonic systems.” Such devices could find applications in biological sensing, ultrafast optics, materials science, and quantum techn

The studies were conducted in collaboration with the Max-Born-Institut im Forschungsverbund Berlin e.V., the NWO Institute AMOLF, the Centro de Fisica de Materiales CSIC/UPV-EHU-Materials Physics Center, and the Donostia International Physics Center (DIPC).?

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