Light-controlled reactions at the nanoscale
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A nanoparticle in the field of a femtosecond laser pulse with tailored waveform and polarization. The controlled enhancement of the field in specific nanoscopic regions of the nanoparticle (yellow spots) induces site-selective photochemical reactions of the molecules adsorbed on the surface. Imaging of the molecular fragments emitted from these regions enables all-optical control of the reaction sites with nanometer resolution. Credit: RMT.Bergues

Controlling strong electromagnetic fields on nanoparticles is the key to triggering targeted molecular reactions on their surfaces. Such control over strong fields is achieved via laser light. Although laser-induced formation and breaking of molecular bonds on nanoparticle surfaces have been observed in the past, nanoscopic optical control of surface reactions has not yet been achieved. An international team led by Dr. Boris Bergues and Prof. Matthias Kling at Ludwig-Maximilians-Universität (LMU) and the Max Planck Institute of Quantum Optics (MPQ) in collaboration with Stanford University has now closed this gap. The physicists determined for the first time the location of light-induced molecular reactions on the surface of isolated silicon dioxide nanoparticles using ultrashort laser pulses.

There is hustle and bustle on the surface of nanoparticles. Molecules dock, dissolve and change their location. All this drives chemical reactions, changes matter and even gives rise to new materials. The events in the nanocosmos can be controlled with the help of electromagnetic fields. This has now been demonstrated by a team led by Dr. Boris Bergues and Prof. Matthias Kling from the Ultrafast Electronics and Nanophotonics Group. To this end, the researchers used strong, femtosecond-laser pulses to generate localized fields on the surfaces of isolated nanoparticles. A femtosecond is one millionth of a billionth of a second.

Using so-called reaction nanoscopy, a new technique recently developed in the same group, the physicists were able to image the reaction site and birthplace of molecular fragments on the surface of silica nanoparticles—at a resolution better than 20 nanometers. The nanoscopic spatial control, achievable at even higher resolution, was brought about by the scientists by superimposing the fields of two laser pulses with different color, and controlled waveform and polarization. Thereby, they had to set the time delay between the two pulses with attosecond accuracy. An attosecond is still a thousand times shorter than a femtosecond. When interacting with this tailored light, the surface of the nanoparticles and the molecules adsorbed there were ionized at targeted sites, leading to the dissociation of the molecules into different fragments.

"Molecular surface reactions on nanoparticles play a fundamental role in nanocatalysis. They could be a key to clean energy production, in particular via photocatalytic water splitting," explains Matthias Kling. "Our results also pave the way for tracking photocatalytic reactions on nanoparticles not only with nanometer spatial resolution, but also with femtosecond temporal resolution. This will provide
detailed insights into the surface processes on the natural spatial and temporal scales of their dynamics," adds Boris Bergues.

The scientists anticipate that this promising new approach can be applied to numerous complex isolated nanostructured materials. Their study is published in Optica.

**More information:** Wenbin Zhang et al, All-optical nanoscopic spatial control of molecular reaction yields on nanoparticles, Optica (2022). DOI: 10.1364/OPTICA.453915

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