Flexible control of light-matter interaction on the nanoscale

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Spatiotemporal control of optical near-fields takes adaptive optics and dynamic-light localization beyond the diffraction limit and potentially into new information-processing devices.

Lasers abound in our daily lives, for example, in compact disk and digital video disk players, pointers, scanners, light shows, and medical or industrial applications. The desired function is achieved by moving light to a certain position at a given time. This light control provides the basis for optical spectroscopy (linear/nonlinear, frequency-/time-resolved), technology (light sources, displays, sensors, markers, solar cells, data storage), and biological processes (photosynthesis, vision). In all of these examples, the temporal and spatial properties of the light field on the relevant time and length scales determine the outcome and thus the desired function, efficiency, and success of the application. Hence, the most general flexibility would be reached if light fields could be created with arbitrary, user-specifiable characteristics in both time and space. In conventional far-field optics, the spot size for such controlled optical interaction is limited by diffraction to about half the wavelength.

We recently demonstrated dynamic positioning of light with ultrafast timing on a target area smaller than the minimum spot size of the laser itself, which is well below the diffraction limit. In effect, this corresponds to the smallest and fastest laser show possible and provides a new and flexible means to control light-matter interaction on the nanoscale. Applications arise in nanotechnology, optical information processing, quantum computation, nanosensing, or other novel microscopy and spectroscopy methods.

Mark I. Stockman, a theoretical physicist at Georgia State University, suggested in 2002 that pulse shaping could have significant uses in the field of nano-optics. In a collaborative effort, Tobias Brixner (University of Würzburg, Germany), F. Javier García de Abajo (Instituto de Optica, Madrid, Spain), and Walter Pfeiffer (University of Bielefeld, Germany) extended his theory and showed—again in a theoretical investigation—that polarization pulse shaping, the ability to switch the polarization state of light within one ultrashort laser of few femtoseconds duration, is essential to achieve simultaneous spatial and temporal control over optical near-field distributions.

Figure 1. Artistic view of spatiotemporal near-field control. An ultra-short polarization shaped light pulse shown in a 3D representation and the switching between different polarization states within about 100fs leads to photoelectron emission (red spheres) from different areas of the sun-shaped nanoantenna (700nm diameter) at different times. Ultrafast variation of wavelength, amplitude, and polarization of the incident laser pulse determines the momentary near-field. The ability to manipulate such excitations opens new possibilities in nanophotonics, and could lead to new concepts for active devices in information processing.

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We extended the collaboration and combined two-photon photoemission electron microscopy and adaptive polarization pulse shaping to demonstrate adaptive near-field control. More recently, we were able to monitor directly the spatiotemporal evolution of a nanophotonic excitation (see Figure 1).

Taking the demonstrated concept of spatiotemporal near-field control to applications requires understanding the underlying mechanisms. The far field that illuminates the nanostructure is composed of two orthogonal polarization components. When illuminating a nanostructure, the local fields generated by these two components are no longer orthogonal, and thus interference effects can be exploited to control the local-field distribution. The phase difference between the two polarization components in the far field is what controls the spatiotemporal field evolution near the nanostructure. Additional control mechanisms, such as matching the incident polarization to oriented dipolar modes of the nanostructure or compensating the spectral phase of the local response, further help to achieve the desired near-field properties.

The emerging field of spatiotemporal nanophotonic control substantially extends the flexibility of light-based manipulation of matter. The ability to generate ultrashort laser pulses revolutionized rather diverse fields including, femtochemistry, material processing, and soft x-ray generation. An additional degree of freedom, such as the spatial control of ultrashort excitation on a sub-diffraction length scale, will also impact many areas, such as biophotonics or quantum nano-optics.

Having now demonstrated nanoscale spatiotemporal near-field control, we are presently developing strategies to apply this scheme in spectroscopy and to try demonstrating new applications. The additional degrees of freedom overcome limitations of conventional far-field excitation, and we plan to apply this flexibility to control the excitation in quantum dot aggregates and semiconductor films with unprecedented spatial and temporal resolution. This will allow the study of nanoscale energy and electron transfer mechanisms with femtosecond time resolution. In the long term, we anticipate that this scheme will find applications in innovative nano-optical quantum computation methods.

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