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Imaging of the Second-harmonic Response of Spatially-oriented Individual Ion-shaped Nanoparticles

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Abstract— During the last decade, many efforts have been made to develop techniques to integrate nanostructures in functional matrices. This activity, mainly boosted by advances in nanofabrication, has enabled the development of elegant methods for the development of planar nanodevices. However, the design and implementation of embedded three-dimensional (3D) nano-architectures with tunable spatial orientation remains a challenge. To overcome this difficulty, an alternative is offered by the technique of sculpturing nanoparticles using ion beams (ion-beam shaping). Here, we use this method to produce an array of anisotropic and spatially-oriented gold nanoparticles embedded in silica matrix. Their orientation is then imaged by nonlinear optical microscopy based on second-harmonic generation and polarized optical beams.

The arrays of gold nanoparticles were fabricated by first preparing an array of spherical particles in silica matrix. These particles were then illuminated by a beam of xenon ions. Depending on the total ion fluence, the gold particle elongate along the direction of irradiation, while maintaining constant volume, allowing nanorods and even nanowires to be fabricated. The tilt angle of the particles was adjusted by the direction of ion irradiation.

Second-harmonic microscopy of nanorods was based on using linearly and radially polarized beams focused by a high-numerical-aperture objective. This technique allows the transverse and longitudinal field components in the focus to be controlled. This in turn affects the coupling of the incident light to the nanorods with different orientations.

We report the high sensitivity of the second-harmonic response to the orientation of the nanorods for different states of polarization. The experimental results were obtained to be in very good agreement with simulations based on the boundary-element method. Compared with previous reports, our results provide a considerable improvement for understanding the interaction of highly focused beams with anisotropic sub-wavelength structures.