Third-harmonic microscopy of Mie-resonant dielectric oligomers

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Nanostructures based on high-index dielectric nanoparticles with optical response governed by multipolar Mie-type resonances has been proved to enhance the efficiency of nonlinear optical effects. Strong local-field confinement in their volume for the magnetic dipolar resonance (MDR) excitation as well as low losses comparing to plasmonic analogues makes them attractive for nonlinear optical applications. Combining dielectric particles in oligomers leads to the collective modes formation governed by near-field coupling between the constituent nanoobjects, that results in largely increased nonlinear optical response. This effect is sensible both for the nanocluster geometry and laser excitation conditions such as pump beam polarization.

In this work we investigated experimentally and numerically the third-harmonic generation (THG) from isolated subwavelength oligomers made of three and four Si nanodisks (trimers and quadrumers) in spectral vicinity of their MDR under different fundamental beam polarizations. We observed a strong dependence of the THG power on the pump beam polarization using nonlinear microscopy setup based on near-infrared femtosecond laser with the ability of the polarization type change (linear polarization (LP), azimuthal polarization (AP) and radial polarization (RP)) and sample azimuthal angle rotation. We demonstrated, that the THG intensity from resonant samples pumped with AP vector beams is seven times larger comparing to the LP pump beams due to the efficient excitation of collective out-of-plane magnetic modes of oligomers by AP light. We also observed the rotational dependence of the THG signal from the clusters pumped by LP beams in accord with their point-group symmetry governed by near-field interaction of nanodisks. Experimental data are in a good agreement with analytical and numerical predictions. Results described above provide a route toward tailoring nonlinear properties of nanoscale Mie-resonant structures and open new functionalities for nanophotonic active devices.