

NONLINEAR PHOTOCHROMIC SWITCHING IN THE PLASMONIC FIELD OF A NANOPARTICLE ARRAY

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Plasmonic nanostructures provide unique environments for non-resonant excitation and switching of photochromic compounds. In this study, photochromic diarylethene molecules were deposited on top of a periodically ordered array of gold nanorods (170 x 80 nm) and then irradiated with <100 fs laser pulses. Irradiation at 800 nm drives the plasmon resonance of the nanoparticle array and induces the photochromic conversion of molecules via non-resonant two-photon excitation. Transmission measurements using broadband continuum laser pulses probe the progress of the photochemical cycloreversion reaction as molecules switch from a visible-absorbing closed-ring structure to a transparent open-ring structure. The spatial dependence of the two-photon conversion of molecules in the plasmonic near field of the array is modeled using calculated field enhancements, and compared with similar measurements for a film of molecules on a glass substrate. Wavelength-dependent polarization effects in the near field of the array lead to interesting anisotropy results in the transmission signal. The results emphasize the importance of both the spatial dependence and anisotropy of the enhanced electric fields in driving non-resonant photochromic reactions.