Ultrafast Dichroism via Photoinduced Symmetry-Breaking in Plasmonic Metasurfaces

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We theoretically predict and show by ultrafast pump-probe experiments a sub-picosecond transient dichroism in a plasmonic metasurface driven by the inhomogeneous space-time dynamics of the photogenerated hot carriers.
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Ultrafast photonics is an emerging field of research which aims at developing devices to modulate light with unprecedented speed [1]. In this framework, the optical nonlinearities of nanostructured media illuminated with intense fs-laser pulses are particularly promising. Plasmonic structures have demonstrated a giant delayed third-order nonlinear response, governed by the internal relaxation of the metallic system [2,3]. Although the ultrafast temporal dynamics following photoexcitation has been extensively investigated, the sub-picosecond spatial inhomogeneities occurring at the nanoscale have been so far neglected. In this work, we theoretically predict and experimentally show that the inhomogeneous space-time distribution of photogenerated hot carriers induces a transient symmetry breaking in a plasmonic metasurface made of highly symmetric metaatoms. Such a fully reversible and ultrafast effect is exploited to address all-optical manipulation of light polarization.

Our concept is described in Figure 1. We predict and observe that the photo-induced spatio-temporal dynamics of hot carriers can break the symmetry of a plasmonic metasurface made of C4-symmetric gold nanocrosses (Fig. 1a), generating a broadband dichroic response with recovery of the isotropic initial state within less than 1 ps. Such effect is indeed much faster than any relaxation (electron-phonon, phonon-phonon) process, as it is governed by electrons thermalisation and diffusion. The polarisation-degeneracy of static transmittance (Fig. 1b) is broken by the inhomogeneous absorption pattern of the pump, locally affecting the metaatoms electronic energy population and generating a non-uniform distribution of out-of-equilibrium carriers. The corresponding metal permittivity changes evolve therefore anisotropically in space (Fig. 1d), inducing a transient transmission anisotropy, revealed by a polarisation-resolved pump-probe ultrafast experiment (Fig. 1c,e). The ultrafast dichroism is modelled via an Inhomogeneous Three-Temperature Model (I3TM) including the spatial dependence. Predictions are then compared with polarisation-resolved experiments using 30-fs pump pulses and probe pulses in the visible range, where a very good agreement is achieved.

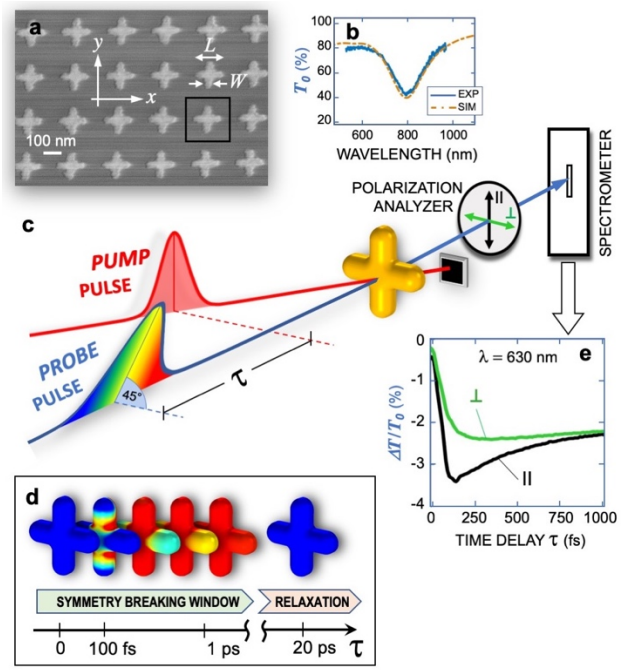

Fig. 1 a. SEM image of the metasurface. b. Measured (blue) and simulated (orange) transmittance of the unperturbed sample. c. Polarisation-resolved pump-probe experimental set-up. d. Transient permittivity pattern (at 630 nm) evolving over time at the nanoscale. e. Differential transmission signal (at 630 nm wavelength) for probe polarisation parallel (black) and orthogonal (green) to the pump.

Our results pave the way for the all-optical control of light polarization at Tera bit/s speed [4], with relevance for telecom applications and novel time-resolved optical spectroscopies.

References

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