Shaping Ultrafast Hot Electrons to Drive Photoinduced Anisotropy in Nonlinear Plasmonic Metasurfaces

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Abstract: We theoretically predict and experimentally demonstrate by transient absorption spectroscopy that spatiotemporal dynamics of hot electrons can promote and control an ultrafast photoinduced anisotropy in plasmonic metasurfaces, enabling active reconfiguration of the nanostructure nonlinear response.

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Photoexcited hot carriers in plasmonic nanostructures hold great promise in a wealth of applications, especially for ultrafast nanophotonics, as they represent an unparalleled channel for enhanced light-matter interactions on the ps timescale [1]. When combined with the flexibility of nonlinear metasurfaces (engineered quasi-2D nanomaterials) in manipulating electromagnetic fields [2], the control of light is achieved at ultrafast speed in ultracompact platforms [3]. An open challenge remains the all-optical light modulation, e.g. of its polarisation, over a broadband bandwidth with sub-ps recovery. In addition, to drive full control of light-matter interaction, active reconfiguration of the modulation is crucial. Here, we combine quantitative modelling and transient absorption spectroscopy to demonstrate the tailoring of light properties through ultrafast spatio-temporal shaping of hot carriers in nonlinear metasurfaces.

The rationale behind our approach [4] is sketched in Figure 1a. We designed and fabricated a metasurface made of a square lattice of Au cross-shaped meta-atoms that, being symmetric, provide isotropic transmittance (T). Resonant absorption of an ultrafast (30-fs) pulse linearly polarised along the cross vertical (0°) arm breaks the system optical symmetry. The induced inhomogeneous near fields promote a non-uniform hot carrier distribution, which locally modifies the metal permittivity. The fingerprint of the ultrafast photoinduced symmetry breaking is a transient anisotropy, revealed in a polarisation-resolved pump-probe experiment, which analyses the 10-fs broadband probe pulses differential transmission $\Delta_{0(90)}=\Delta T_{0(90)}(\lambda,\tau)/T(\lambda)$ along the vertical (0°) and horizontal (90°) directions. The metric $\Delta_{90} - \Delta_0$ measures the hot carrier-driven ultrafast anisotropy and indicates a broadband linear dichroism (Fig. 1b) vanishing in 1 ps. Recovery occurs long before relaxation, as the symmetry-breaking window is governed by electron spatial homogenization within meta-atoms.

Moreover, the spatial pattern of the hot-carrier population ultimately stems from the excitation conditions. The pump polarisation establishes the symmetry of the perturbation, acting as a degree of freedom so far unexplored for all-optical reconfiguration [5]. By rotating the polarisation direction, the photoinduced transient dichroism is readily controlled in intensity, up to full switch in the dichroism sign for two pump pulses differing by 90°. Theoretical predictions of such sign reversal are demonstrated by ultrafast experiments, proving all-optical reconfiguration dictated by hot carrier inhomogeneities at the nanoscale (Fig 2c).

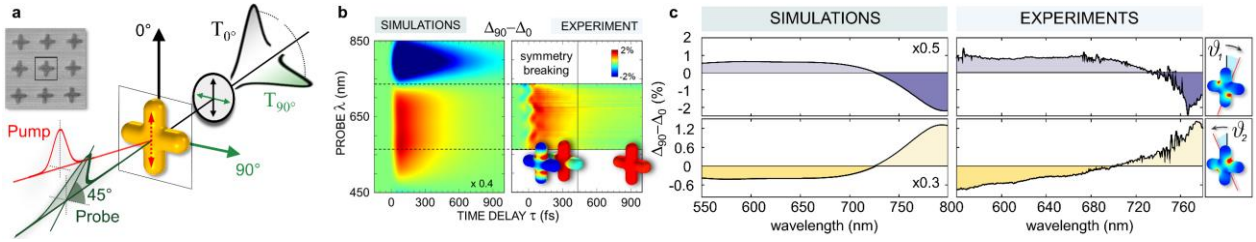

Fig. 1. a) Sketch of the polarisation-selective transient absorption set-up and SEM image of the sample. b) Simulated and measured sub-ps broadband dichroism, where the photoinduced symmetry breaking window is highlighted. c) Simulated and measured spectra of the dichroic signal at 100 fs for a pair of pump pulses with polarisation angles (ϑ_1 and ϑ_2) differing by 90°.

In conclusion, we have demonstrated that photoexcitation of a plasmonic metasurface by an ultrashort optical pulse induces spatial inhomogeneities of hot carriers, driving an ultrafast anisotropy in an otherwise isotropic metasurface. Modulation, governed by the spatial distribution of photoexcited electrons, is tailored by varying the excitation conditions. Full reconfiguration, up to sign reversal, is achieved by tuning the pump polarisation direction. Our approach paves the way to all-optical manipulation of light at Terabit/s speed and suggests a route for actively tailoring the control in a flexible, fully all-optically reconfigurable platform based on nonlinear metasurfaces.

References

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We acknowledge the METAFAST project (no. 899673) funded by the EU H2020 Research and Innovation programme.