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Photoinduced transient symmetry breaking in plasmonic structures for ultrafast nanophotonics

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ABSTRACT

We study the spatio-temporal evolution of hot electrons generated in plasmonic nanostructures under resonant excitation with fs-laser pulses. A spatially inhomogeneous version of the Three-Temperature Model for hot-electrons dynamics, coupled to semiclassical calculations of third-order optical nonlinearity in gold, enabled us to engineer a transient symmetry breaking of the optical properties at the nanoscale. This effect is exploited to achieve all-optical control of light with unprecedented speed. For instance, a photoinduced broadband dichroism, fully reversible and transiently vanishing in less than 1 picoseconds (overcoming the speed bottleneck caused by slower, electron-phonon and phonon-phonon relaxation processes), has been experimentally demonstrated in plasmonic metasurfaces with nanocross metaatoms. Also, we designed a nonlinear plasmonic metagrating (based on cross-polarized gold nanostrip dimer metaatoms), where the nanoscale symmetry breaking enables ultrafast reconfiguration of diffraction orders via control laser pulses. The photoinduced power imbalance between symmetrical diffraction orders is calculated to exceed 20% under moderate (~2 mJ/cm^2) laser fluence, and returns to the balanced diffraction in about 2 ps. Our design has been developed for gold nanomaterials, but the concept of ultrafast all-optical symmetry breaking can be exploited beyond plasmonics (e.g. in semiconductor nanostructures), with potential impact on a broad range of applications in nanophotonics.

Keywords: Hot electrons; plasmonic structures; metasurfaces; pump-probe spectroscopy; nonlinear optics; polarization control; diffraction management

1. INTRODUCTION

Hot electrons in plasmonic nanostructures have been the subject of intensive research, with particular interest to the temporal dynamics following excitation with femtosecond laser pulses (see e.g. Ref. 1 for an overview). However, the effects linked to spatial local inhomogeneities of hot electrons have been overlooked until very recently [2-7].

Here, we show that the resonant excitation of highly symmetric plasmonic nanostructures can induce an ultrafast photo-thermal symmetry breaking, governed by the spatial inhomogeneities of the photogenerated hot electrons. This effect can be exploited in ultrafast nanophotonics to achieve unprecedented functionalities from planar periodic configurations, i.e. plasmonic metasurfaces. Actually, optical metasurfaces can strongly enhance nonlinear effects and make it possible to reconfigure their optical response using ultrashort laser pulses [8-10]. In particular, plasmonic metasurfaces have turned out to be promising because of the giant third-order nonlinearity of noble metals, which is governed by photogenerated hot carriers [11-14].

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2. RESULTS AND DISCUSSION

2.1 Ultrafast Broadband Dichroism in Plasmonic Metasurfaces

We designed a plasmonic metasurface (square lattice arrangement with ~270-nm periodicity) of C4-symmetric gold nanocrosses with 45-nm thickness, 60-nm width and 165-nm length of the nanocross arms (Fig. 1a).

The metasurface was fabricated by electron beam lithography on a transparent substrate (CaF_2) [6].
Due to its high symmetry, this nanomaterial provides a polarization-independent static transmittance at normal incidence, characterized by a broad extinction peak at 800 nm, arising from the degenerate longitudinal plasmonic resonances of the two arms. Such a degeneracy can be broken by the resonant absorption of an ultrashort control pulse with linear polarization parallel to the direction of one of the arms (Fig. 1a).

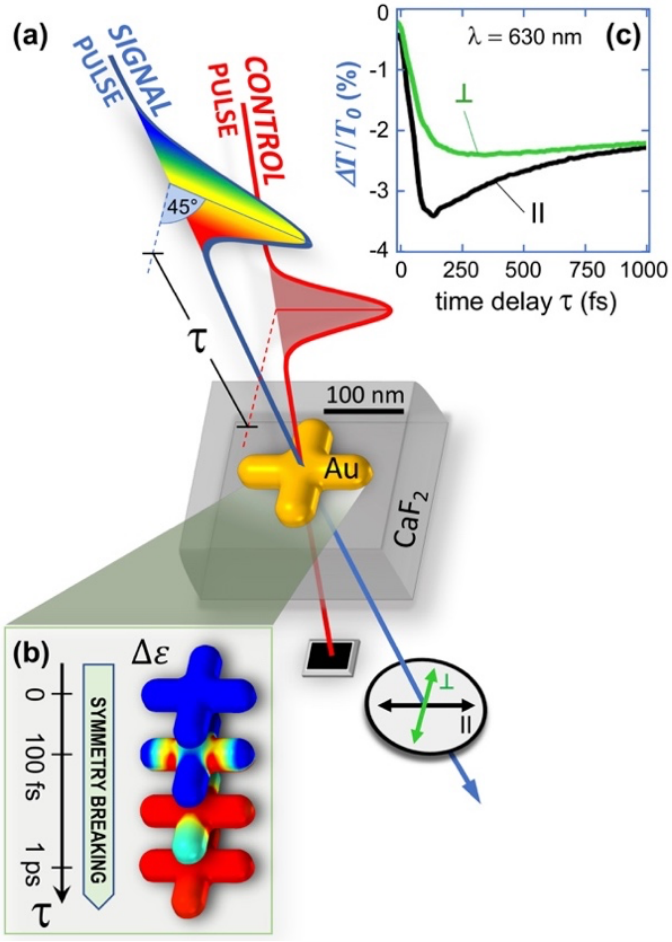

Figure 1. (a) Experimental setup for ultrafast transient dichroism. A sketch of the Au-nanocross metaatom supported on a dielectric substrate is shown. The meta-atom is excited with ~30-fs control laser pulses at 860 nm (red), generating hot electrons at the nanoscale. (b) The C4 symmetry of the nanocross is broken by the highly inhomogeneous spatial pattern of the photoinduced permittivity modulation on the sub-picosecond time scale. (c) Polarization-resolved analysis of the transmitted signal pulse at 630 nm wavelength impinging on the metasurface at a time delay τ with respect to the control pulse (with ~400 μJ/cm^2 fluence).

Indeed, photoexcitation creates a highly inhomogeneous near field, mostly because of the retardation-based nature of plasmonic modes in relatively large nanostructures.
The inhomogeneous absorption pattern in each meta-atom locally affects the electronic energy distribution of gold, inducing a non-uniform out-of-equilibrium hot-carrier distribution that anisotropically modifies the metal permittivity on

a time scale of 1 ps (Fig. 1b). The fingerprint of the ultrafast photoinduced symmetry breaking is revealed as a transient transmission anisotropy from polarization-resolved pump-probe measurements (Fig. 1c). Note that the degeneracy between the two polarizations is restored in about 1 ps, i.e. much earlier than the onset of electron-phonon relaxation processes (taking place on the time scale of 10 ps) [6].

2.2 All-optically reconfigurable plasmonic metagrating

By exploiting the same approach, we designed a plasmonic metagrating with 800 nm periodicity and unit cell made of a bent gold nanostrip supported on a sawtooth CaF_2 substrate (Fig. 2a). A p-polarized ultrashort laser pulse at 600 nm shined at 45° angle of incidence is capable of inducing a highly inhomogeneous spatial pattern of the photogenerated hot electrons.

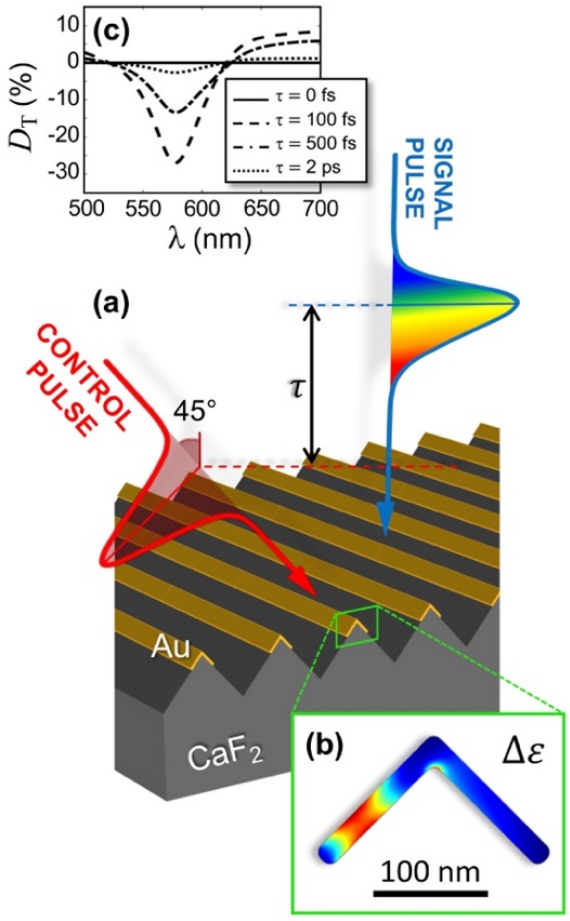

Figure 2. (a) Cartoon of the all-optically reconfiguration plasmonic metagrating for ultrafast diffraction management via absorption of visible control laser pulse (50 fs duration) impinging at 45° angle of incidence. (b) Highly inhomogeneous permittivity modulation pattern following photoinduced hot electrons generation. (c) Transient differential impairment between +1 and -1 transmission orders for a broadband signal pulse interacting with the metagrating at a time delay τ with respect to a control pulse of ~2 mJ/cm^2 fluence.

The subsequent nonlinear permittivity change thus breaks the left-right symmetry of the cell (Fig. 2b) and causes a transient imbalance between symmetric diffraction orders, for a broadband signal pulse impinging at normal incidence [7]. The effect is measured in terms of the D_T figure of merit (Fig. 2c), defined as follows:

$$D_T(\lambda,\tau) = \frac{T_{+1}(\lambda,\tau) - T_{-1}(\lambda,\tau)}{2T(\lambda)}.$$

In above formula, $T_{+1}(\lambda,\tau)$ and $T_{-1}(\lambda,\tau)$ are, respectively, the dynamic +1 and -1 transmission spectra, and $T(\lambda)$ is the static (and degenerate) +1/-1 transmission. Note that thanks to the high thermal conductivity of the metal, causing high speed homogenization of electronic temperature, the degeneracy between +1 and -1 orders is restored in about 2 ps [7].

3. CONCLUSIONS

We demonstrated the ability to produce a transient optical symmetry breaking in plasmonic metastructures via photoinduced hot electrons. The associated thermo-modulational nonlinearity is exploited to achieve unprecedented control over light beams, including ultrafast polarization switching and ultrafast diffraction management from planar plasmonic metastructures. We also envisage potential application of our results to hot-electron harvesting, for the optimization of plasmon-enhanced photocatalysis and photodetection configurations thanks to a refined design, properly accounting for short-lived nanoscale inhomogeneities.

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