# Giant Nonlinearity of an Optically Reconfigurable Plasmonic Metamaterial

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# Giant nonlinearity of optically reconfigurable plasmonic metamaterial

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**Keywords:** optical nonlinearity, plasmonic nanostructures, reconfigurable metamaterials, metasurfaces

From the demonstration of saturable absorption by Wawilow and Levshin in 1926,[1] and with invention of the laser, unavailability of strongly nonlinear materials was a key obstacle for developing optical signal processing, in particular in transparent telecommunication networks. Today, most advanced photonic switching materials exploit gain dynamics[2] and near-band and excitonic effects in semiconductors,[3] nonlinearities in organic media with weakly-localized electrons[4] and nonlinearities enhanced by hybridization with metamaterials.[5] Here we report on a new type of artificial nonlinearity that is nano-opto-mechanical in nature. It was observed in an artificial metamaterial array of plasmonic meta-molecules supported by a flexible nano-membrane. Here nonlinearity is underpinned by the reversible reconfiguration of its structure induced by light. In a film of only 100 nanometres thickness we demonstrate modulation of light with light using milliwatt power telecom diode lasers.

Some exceptional opportunities for developing engineered nonlinear media are provided by reconfigurable nanostructures that take advantage of the changing balance of forces at the nanoscale.[6-9] With the decrease in the physical dimensions of a system the electromagnetic forces between constituent elements grow, as may be illustrated by the repulsion of electrons as their separation diminishes. In contrast, elastic forces, such as the force restoring a compressed spring, decrease with size. Moreover, the nanoscale metamaterial building blocks have high natural frequencies and thus can be moved very fast, potentially offering GHz switching bandwidth for elements of sub-micron size.

Recently, this was exploited to develop reconfigurable metamaterials fabricated on nanoscale elastic membranes. These photonic metamaterials can be driven thermo-elastically[10] and with electromagnetic forces, such as the Coulomb force between charged elements of the nanostructure,[11] or the Lorentz force acting on currents running through conductive elements in magnetic field.[12] Their electro-optical and magneto-optical switching characteristics surpass those of natural media by orders of magnitude.

Metamaterials are in essence arrays of optical resonators where light-induced nanomechanical phenomena can play crucial roles. For instance it has been suggested that nearfield[13] and Casimir forces[14,15] in metamaterials are modified in the presence of light. Forces between oscillating plasmonic or displacement currents induced by light in metamolecules were also theoretically shown to be sufficient to drive reconfiguration of a metamaterial structure in the optical part of the spectrum,[16-18] and proof-of-principle experiments on such interactions between individual metamolecules have been reported at microwave frequencies.[19]

Here we show that reversible reconfiguration of a plasmonic metamaterial nanostructure driven by optical forces between its illuminated elements can be the source of a strong optical nonlinearity. We show that this cubic nonlinearity may be used to modulate light with light at milliwatt power levels. Moreover, although for the majority of media the magnitude of nonlinearity tends to be proportional to its response time, the nano-opto-mechnical nonlinearity is three orders of magnitude faster than could be expected from this otherwise universal trend.

The plasmonic nanomechanical metamaterial was fabricated on strips of suspended dielectric membrane of nanoscale thickness in such a way that plasmonic elements of the metamolecules were located on different strips. Here light illumination leads to electromagnetic and thermal forces that induce nanoscale strip displacements and thus reconfiguration of individual metamolecules in a way that affects their plasmonic resonances, see **Figure 1**. This leads to modulation of the metamaterial's transmissivity at different wavelengths. The nonlinear response of metamaterial, that is only 100 nanometres thick, can be observed at only a few milliwatts of continuous laser power, using lasers operating at telecommunication wavelengths.

We developed optically reconfigurable metamaterial on the basis of a Π-shaped resonator design known for exhibiting plasmon-induced transparency,[20,21] see **Figure 1** and Supplementary **Figure S1a**. To allow mechanical deformation of the 700 nm × 700 nm plasmonic Π metamolecules, the horizontal and vertical gold bars were supported by different flexible silicon nitride strips of 28 μm length spaced by alternating gaps of 95 nm and 145 nm. The nanostructure was fabricated by focused ion beam milling from a 50 nm thick silicon nitride membrane covered by 50 nm of gold. Such structures can be reconfigured by (i) differential thermal expansion through light-induce heating of the bimorph gold and silicon nitride layers and (ii) by near-field optical forces acting between elements of illuminated plasmonic resonators.

Experimental measurements and full 3D Maxwell simulations show a pronounced near-infrared absorption resonance around 1240 nm, see **Figure 2a** and Supplementary **Figure S1b**. Maxwell stress tensor calculations[16] assuming normal incidence illumination of the non-diffracting, periodic metamaterial reveal optical forces acting on the Π-resonators around these resonances, see **Figure 2b-c**. As the normally incident photons only carry momentum along the $z$-direction the net force along $z$ must comply with the momentum transfer associated with absorption $A$ and reflection $R$, $F_{z1}+F_{z2}=(A+2R)P/c$, where $P$ is the incident power per unit cell area and $c$ is the speed of light in vacuum. In close agreement with these relationships, our simulations show substantial differential optical forces $F_2-F_1$ between the unit cell's strip segments reaching about 0.4 $P/c$ along $y$ and 2.8 $P/c$ along $z$.

Optical forces in such metamaterial nanostructures can be understood as time-averaged Coulomb and Lorentz forces acting between the currents and oscillating dipole charges of the plasmonic resonators in the presence of the illuminating electromagnetic wave.[18] For example, repulsive and attractive optical forces within the metamaterial plane for illumination at wavelengths of 1310 nm and 1550 nm correspond to repulsive and attractive interaction of the dipole charges on neighboring strips, see **Figure 2a-b**. Forces normal to the metamaterial plane result from a combination of optical pressure and the Lorentz force acting on the moving charges of the plasmonic mode in the presence of the incident wave's magnetic field.

We investigated the nonlinear optical properties of the metamaterial in a pump-probe experiment as presented in **Figure 3a**. As the pump and probe optical sources we used fiber-coupled telecommunication laser diodes operating at the wavelengths of 1550 nm and 1310 nm, respectively. The intensity of the pump beam was modulated by a fibre-coupled electro-optical modulator. The pump and probe were then combined into a single beam using a fiber coupler, de-coupled into free space and focused on the sample placed in a microscope using focusing and collection objectives. The sample was placed in a cell evacuated to the pressure of 30 Pa to prevent damping of the nanomechanical motion. Light transmitted through the sample was detected by an InGaAs photodetector and a lock-in amplifier locked to the pump modulation frequency. In order to optimize the light-induced nano-opto-mechanical modulation, we pumped the nanostructure at 1550 nm just above its absorption resonance, where simulations predict significant in-plane forces.

At low modulation frequencies, below 100 kHz, the optical pump leads to pronounced modulation of the metamaterial's transmission at the probe wavelength, see **Figure 3c**. For 0.9 mW pump power (peak intensity 9.2 μW μm$^{-2}$) a modulation amplitude on the order of 1% is observed at 25 kHz modulation. This is an exceptionally large effect considering that the interaction length of light and metamaterial is less than one tenth of a wavelength. In contrast, in conventional nonlinear optical media interaction lengths much larger than the wavelength are required to accumulate significant effects. The low-frequency component of the nonlinear response has a thermal nature and drops rapidly with increasing modulation frequency, fading at a few 100s of kHz. In this frequency range, the modulation amplitude halves as the modulation frequency doubles which is consistent with a thermal deformation of the bimorph Au/SiN$_x$ membrane, see inset to **Figure 3b**. This is consistent with thermal calculations for 50 nm gold[22] and silicon nitride[23] layers, which predict conductive cooling timescales on the order of 20-30 μs. The light-induced heating of the nanostructure is balanced by heat conduction along the metamaterial strips and according to our calculations can reach hundreds of degrees with pump excitation at 10 μW μm$^{-2}$. As the thermal expansion coefficient of gold ($14.4 \times 10^{-6}$ K$^{-1}$) exceeds that of silicon nitride ($2.8 \times 10^{-6}$ K$^{-1}$) 5-fold such large temperature changes will bend the strips. When heated by the modulated pump laser, narrow strips will rise higher than wider strips due to more complete gold coverage; the metamolecule changes its shape and therefore its plasmonic absorption and transmission for the probe wavelength change.

While the thermal nonlinearity easily dominates at low modulation frequencies, other modulation mechanisms are at play at higher frequencies. Indeed, as differential thermal expansion in the layered structure can only drive motion out of the metamaterial plane, it cannot drive in-plane resonant oscillations of the metamaterial strips. Several resonant peaks of nonlinear response are seen at much higher frequencies around 600 kHz, 1 MHz and 2 MHz, see **Figure 3c**. Mechanical eigenmode calculations identify the resonances as the fundamental modes of oscillation of the narrow and wide strips, corresponding to vibration normal to the metamaterial plane (around 600 kHz) and motion within the metamaterial plane (at MHz frequencies), see insets. Comparing the sub-$\mu s$ heating and cooling cycles to the characteristic strip cooling timescale, we argue for their non-thermal origin.

To assess the origin of the high frequency modulation we estimate the optomechanical deformation that can be expected from the optical forces acting between different parts of the metamolecular resonator. Based on the results of simulations presented in **Figure 2c**, pump-induced optical forces reach 0.5 $P/c$ ( 0.2 $P/c$) for the narrow (wide) strips. For a strip pair with 29 unit cells and 9.2 μW $\mu m^{-2}$ pump intensity, this corresponds to about 220 fN (90 fN) force per strip. Estimating the spring constant $k$ from strip mass $m$ and measured resonance frequency $f$ as $k=(2\pi f)^2\ m$ =28 fN $pm^{-1}$ (56 fN $pm^{-1}$) for out-of-plane deformation, we may expect the gap between strips and thus between elements of the metamolecule to change by 6 pm. Corresponding estimates for in-plane deformation predict a gap change on the order of 1 pm. Such modulation is not sufficient to create a detectable level of non-resonant high-frequency modulation. However, much larger displacements are expected at the structure's mechanical resonances, where the displacement will be enhanced by the quality factor of the mechanical resonator. Our optical measurements show mechanical resonance quality factors on the order of 100, which may serve as a lower limit for the quality factor of mechanical resonances of individual strips as the overall resonance profile is affected by inhomogeneous broadening due to slight structural variations from strip to strip. Taking this enhancement into account, optical forces are sufficient to induce resonant strip displacements on the order of 1 nm, which is consistent with the observed resonant transmission modulation on the order of 1%. Therefore we argue that the resonant response at 600 kHz, 1 MHz and 2 MHz is predominantly driven by non-thermal, optical forces.

Absorption in a nonlinear medium is conventionally described by the expression $-\frac{dI}{dz} = \alpha I + \beta I^2 + \ldots$ , where $I$ is light intensity, $z$ is the propagation distance and $\alpha$ and $\beta$ are the linear and nonlinear absorption coefficients. As the observed nonlinear transmission change is proportional to the pump power, we can quantify the nonlinearity of the reconfigurable photonic metamaterial by estimating its first nonlinear absorption coefficient $\beta$. Assuming that the nonlinear transmission change $\Delta T$ results from nonlinear absorption, $\beta \sim \Delta T/(It)$, where $t$ is the metamaterial's thickness. At 1 MHz modulation frequency, $\beta \sim 5\times10^{-3}$ m W$^{-1}$ corresponding to a nonlinear susceptibility on the order of $\mathrm{Im}(\chi^{(3)})/n^2 \sim 10^{-12}$ m$^2$ V$^{-2}$ and a nonlinear imaginary refractive index change $\Delta\kappa \sim 5\times10^{-3}$ at 9.2 μW μm$^{-2}$ pump intensity. We argue here that the observed nano-opto-mechanical nonlinearity is special as it exhibits a departure from the prevailing relation between speed and magnitude of nonlinear responses (**Figure 4**). This happens in a way that allows larger nonlinearity or higher modulation frequency to be achieved than could be expected from traditional media. Indeed, it appears that for many mechanisms of optical nonlinearity the achievable modulation frequency $v$ is related to the magnitude of the nonlinerity $\chi^{(3)}$ in such a way that $\chi^{(3)}v \sim 10^{-9}$

$m^2V^{-2}s^{-1}$.[24] At the resonance of in-plane motion, MHz rate modulation can be achieved at about 10 µW µm$^{-2}$ of light intensity corresponding to about $\chi^{(3)}\nu \sim 10^{-6}$ m$^2$ V$^{-2}$ s$^{-1}$.

The magnitude of the observed nonlinearity is controlled by the magnitude of the light-induced strip displacement and the displacement sensitivity of the metamaterial's optical properties. Therefore, the observed nonlinearity may be increased further by optimizing the design of the meta-molecules to achieve higher quality factor optical resonances and stronger electromagnetic coupling between the moving parts of the unit cell at the pump and near the probe wavelengths. Higher quality factor optical resonances exhibit higher sensitivity to perturbations as well as increased optical forces and they may be achieved by substituting gold for a lower loss plasmonic material, such as silver, or an almost lossless high index dielectric as suggested recently.[16] Stronger electromagnetic coupling between the moving parts of the unit cell may also be achieved by reducing their spacing. Furthermore, longer strips will increase the magnitude of the nonlinearity by increasing the light-induced displacement, but will also have lower mechanical resonance frequencies due to a reduced spring constant.

Larger modulation amplitudes will not only result from nanomechanical structures with higher nonlinearity (as detailed above), but also from increasing the pump power (**Figure 3**). Thus, the achievable modulation amplitude for a given structure is limited by its thermal damage threshold. For slow modulation – relative to the structure’s cooling timescale – thermal damage is controlled by the peak intensity, while thermal damage for fast modulation is controlled by the average intensity, thus allowing for a two times higher peak intensity. Therefore, we expect that the resonant high frequency modulation in our experimental structure can be increased to several percent by increasing the pump power.

In summary, we provide the first experimental demonstration of a giant nano-opto-mechanical nonlinearity in plasmonic metamaterial that allows modulation of light with light at MHz frequencies and milliwatt power levels. Bimorph deformation resulting from optical heating at low modulation frequencies and electromagnetic forces between elements of plasmonic resonators drive the nonlinear response. The magnitude and spectral position of the underlying optical resonance can be controlled across the optical spectral range by design of the plasmonic resonators and the supporting nano-membrane strips.

## Experimental Section

*Plasmonmechanical metamaterial fabrication*: Starting with a commercially available 50 nm thick low stress silicon nitride membrane, a 50 nm thick gold layer for the plasmonic metamaterial was thermally evaporated. The gold-coated membrane was structured with a focused ion beam system (FEI Helios 600 NanoLab). The pattern of plasmonic resonators was milled and then the supporting silicon nitride membrane was cut into suspended strips with tapered ends. Detailed dimensions of the nanostructure are given by Supplementary **Figure S1b**.

*Experimental characterization*: The cubic nonlinearity was measured by detecting the pump-induced modulation of the metamaterial's transmission (Figure 3a). The probe beam at wavelength 1310 nm was generated by a fiber-coupled telecom laser diode (Thorlabs FPL1053S). The pump beam from a fiber-coupled laser diode at wavelength 1550 nm (Thorlabs FPL1009S) was modulated with a fiberized electro-optical modulator (EOSpace AX-0K5-10-PFA-UL) and then combined with the probe beam in a fiber coupler (JDSU L2SWM1310/1550BB). The transmitted beam was filtered to remove the pump and the probe signal was detected by an InGaAs photodetector (New Focus 1811) and a lock-in amplifier

(Stanford Research SR844). In all optical experiments and simulations the electric field was *x*-polarized (parallel to the strips) and incident on the silicon nitride side of the sample.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the authors.

**Acknowledgements**

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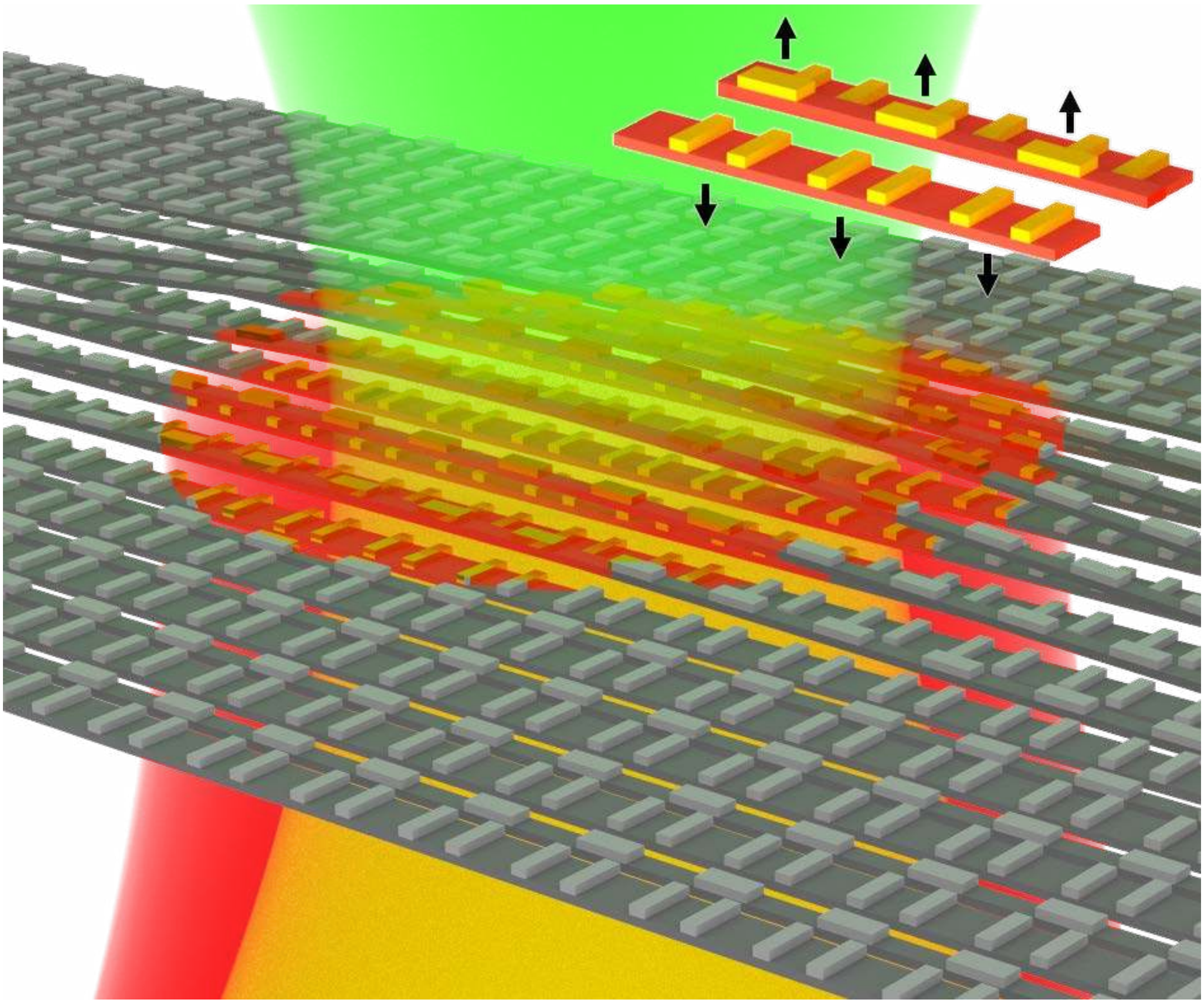

**Figure 1**. Nonlinearity in optically reconfigurable metamaterial. Light-induced (red) forces in the plasmonic metamaterial array cause nanoscale reversible displacements of its small and light building blocks supported by an elastic nano-membrane. They move fast, with a response time reaching microseconds. These displacements change the plasmonic spectra of the metamaterial array and its transmission. This is used to modulate another, weaker beam of light (green) at a different wavelength.

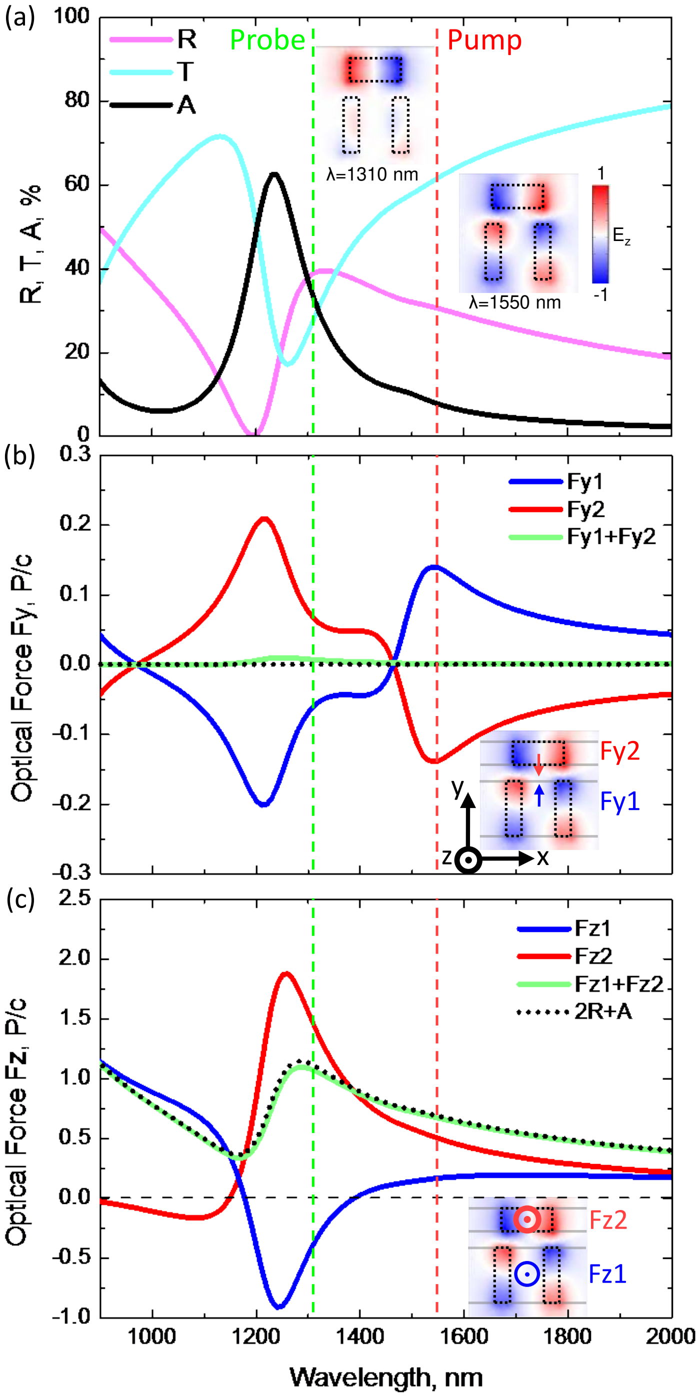

**Figure 2.** Optical spectra and optical forces. (a) Simulated metamaterial transmission *T*, refection *R* and absorption *A* spectra. Insets show maps of the optically induced charge distributions at the probe and pump wavelengths in terms of the instantaneous electric field $E_z$ that these charges generate normal to the metamaterial surface. The maps are normalized to the maximum of $E_z$. (b) In-plane-of-metamaterial component of optical forces $F_{y1,2}$ acting

between the strip segments of an individual unit cell according to Maxwell stress tensor calculations. (c) Normal to the metamaterial component of optical forces $F_{z1,2}$ acting on the strip segments along the light propagation direction. The total optical force on the unit cell (green line) is presented alongside the value expected from reflection and absorption (dotted black line). Forces are shown per unit cell in units of *P/c*, where *P* is the incident power per unit cell and *c* is the speed of light in vacuum. All quantities are shown for *x*-polarized illumination of the silicon nitride side.

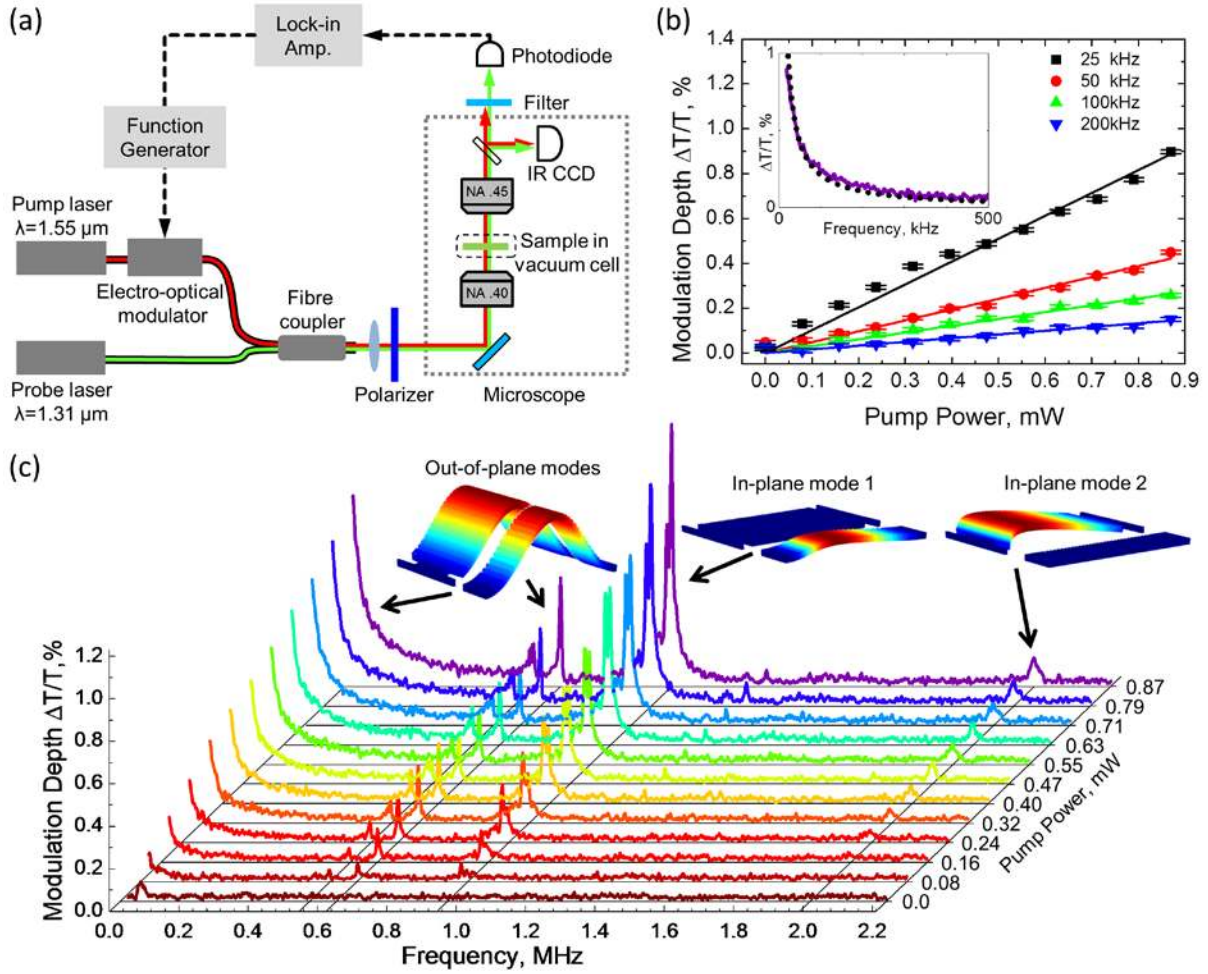

**Figure 3.** Modulating light with light using optically reconfigurable metamaterial. (a) Schematic of the pump-probe experimental setup. (b) Probe transmission modulation depth at $\lambda$=1310 nm as a function of pump power for several pump modulation frequencies with linear fits (solid lines). The inset shows a hyperbolic fit ~*1/f* (black dotted line) of the thermal modulation tail for a pump power of 0.9 mW. (c) Modulation depth as a function of pump laser power and modulation frequency. Corresponding mechanical modes of the metamaterial nanostructure are shown near resonant peaks. The pump modulation depth is close to 100% in all cases.

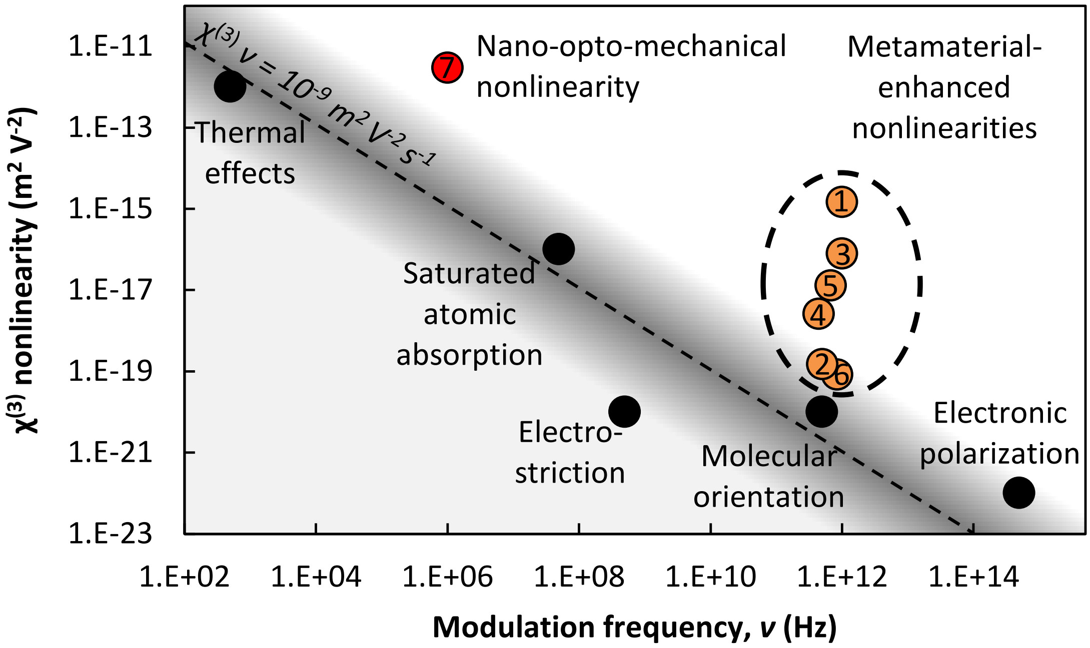

**Figure 4.** Optical nonlinearities in metamaterials and conventional media. Cubic optical nonlinearities due to thermal effects, saturated absorption, molecular orientation and electronic anharmonicity follow a general trend of decreasing achievable modulation frequency $v$ with increasing magnitude of the nonlinearity: $\chi^{(3)}\ v \sim 10^{-9}\ \mathrm{m}^2\mathrm{V}^{-2}\mathrm{s}^{-1}$ .[24] Metamaterial patterning of plasmonic metal[25, 26] (points 1 and 2), hybridization of carbon nanotubes[27] (point 3), graphene[28] (point 4), and semiconductors[29, 30] (points 5 and 6) with metamaterials resonantly enhances the magnitude of the nonlinear response without affecting its response time. Optically reconfigurable metamaterial demonstrated in the present work also departs from the common trend exhibiting $\chi^{(3)}v$ that is three orders of magnitude bigger than in conventional nonlinear media (point 7).

**Metamaterial nanostructures actuated by light give rise to a large optical nonlinearity.** Plasmonic meta-molecules on a flexible support structure cut from a dielectric membrane of nanoscale thickness are rearranged by optical illumination. This changes the optical properties of the strongly coupled plasmonic structure and therefore results in modulation of light with light.

**Keywords:** optical nonlinearity, plasmonic nanostructures, reconfigurable metamaterials, metasurfaces

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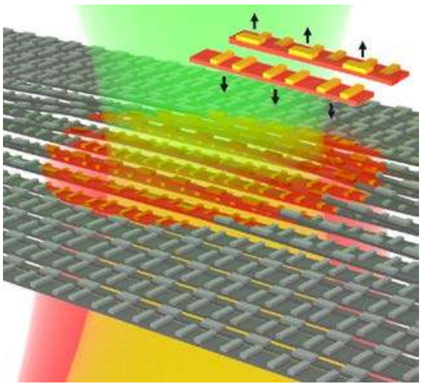
**Giant nonlinearity of optically reconfigurable plasmonic metamaterial**



# Supporting Information

## Giant nonlinearity of optically reconfigurable plasmonic metamaterial

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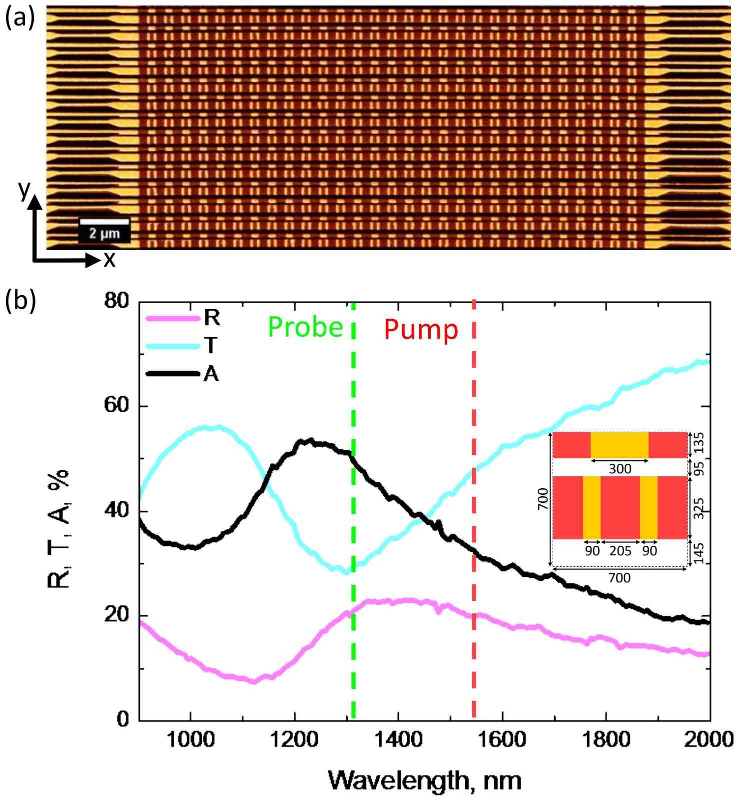

**Figure S1.** Optically reconfigurable plasmonic metamaterial. (a) Scanning electron microscope image of the metamaterial nanostructure consisting of asymmetrically spaced gold (yellow) resonators on silicon nitride strips (red). (b) Transmission T, refection R and absorption A spectra of the metamaterial for x-polarized light incident on the silicon nitride side. Dashed lines indicate the optical 1310 nm probe and 1550 nm pump wavelengths used in the modulation measurements of the main manuscript. The inset shows detailed unit cell dimensions in nanometers. The scanning electron micrograph was recorded by FEI Helios 600 NanoLab and the optical spectra were measured using a microspectrophotometer (CRAIC Technologies).