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Large enhancement of third-order nonlinear effects with a resonant all-dielectric metasurface

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A novel low-profile nonlinear metasurface, consisting of a single-layer of all-dielectric material, is proposed and numerically investigated by a nonlinear full-wave finite-difference time-domain (FDTD) method. The proposed metasurface is transparent for low, and opaque for high values of incident light intensity. The metasurface design is broadly applicable to enhancement of intrinsic nonlinearities of any material with a sufficiently high refractive index contrast. We illustrate the ability of this design to enhance intrinsic nonlinear absorption of a transition metal oxide, vanadium pentoxide (V_2O_5), with resonant metasurface elements. The complex third-order nonlinear susceptibility $\left(\chi^{(3)}\right)$ for V_2O_5, representing both nonlinear refraction and absorption is considered in FDTD simulations. Our design achieves high initial transparency (>90%) for low incident light intensity. An order of magnitude decrease in the required input light intensity threshold for nonlinear response of the metasurface is observed in comparison with an unpatterend film. The proposed all-dielectric metasurface in this work is ultrathin and easy to fabricate. We envision a number of applications of this design for thin film coatings that offer protection against high-power laser radiation. [http://dx.doi.org/10.1063/1.4967818]

I. INTRODUCTION

Nonlinear materials that can limit transmission of laser pulses on sub-nanosecond time scale are of interest for communications,[1] as well as for protection of eyesight and sensitive devices from intense laser pulses.[2,3] In order to realize fast switching speeds, materials with nonlinearities based on electronic transitions are desirable. Such materials may include carbon allotropes, nonlinear dyes,[4] semiconductors irradiated at photon energies below bandgap,[5,6] and plasmonic nanoparticles.[7] While considerable research on these materials has been performed to date, lowering the onset of nonlinear threshold with sufficiently high damage resistance still remains elusive.

One strategy to increase intrinsic material nonlinearities is through nanoresonator designs that can considerably enhance incident field on nanoscale, thereby increasing the local field intensity. Such a field localization approach has been previously demonstrated with plasmonic nanostructures. In that work, by pattering a gold film on the nanoscale, a 100-fold enhancement of nonlinear absorption coefficient was demonstrated with femtosecond response time.[8] While this approach seems intriguing, plasmonic materials are prone to damage at higher laser intensities due to a large linear absorption coefficient of metals.[7] For increased initial transparency, 1D photonic crystals with embedded defect

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layers have been proposed as a means of enhancing nonlinearities by field localization in the nonlinear defect layer.[9,10]

Alternatively, the authors have previously reported an ultra-thin metasurface made of a single layer of all-dielectric disks to manipulate optical wavefront by engineering the electric and magnetic dipole modes.[11] Dielectric metasurfaces rely on high refractive index contrast and have substantially lower ohmic losses compared to their plasmonic counterparts. Moreover, in dielectric metasurfaces, resonant fields are localized inside the nonlinear material, contrary to plasmonics metasurfaces that mostly have the fields localized between the adjacent inclusions. This makes dielectric metasurface an ideal candidate for enhancing the nonlinear response of nanostructures with intrinsic nonlinearities.

When considering enhancement of intrinsic nonlinearities while minimizing damage, it is important to choose a starting material with a low linear absorption, high nonlinear absorption, and a high linear refractive index for effective resonant enhancement. In that regard, transition-metal oxides have previously shown, both theoretically[12] and experimentally,[13,14] to possess a large third-order nonlinear optical response. A number of these materials have high linear refractive index n>2. Moreover, stability, strength, and fast response of these compounds make them candidates of high-potential for a body of optical applications.[13,14]

In particular, vanadium pentoxide (V_2O_5) is a transition-metal oxide with centrosymmetric (orthorhombic) lattice structure, which shows a large third-order nonlinear susceptibility and a response time on sub-nanosecond scale.[13,14] The nonlinear absorption coefficient (β) of a 150 nm thick V_2O_5 film is found to be about 3.5×10^{-5} cm/W, that is twice greater than Fe_2O_3 and CuO films.[14] Unlike VO_2, the higher oxidized phase of vanadium dioxide, V_2O_5, is transparent above 500 nm wavelength and has been found to be thermally stable for temperatures as high as 500° C.[15]

Our goal in this letter is to realize a low profile all-dielectric metasurface to protect sensitive optical devices against high-power laser radiation. To achieve this purpose, we demonstrate our novel design based on V_2O_5 as the building material of the metasurface. Here, we consider a periodic array of air holes in a V_2O_5 film. As an irradiation geometry, we consider an incident laser beam that becomes focused by the first lens at the position of the metasurface (Fig. 1). This lens is needed to bring the intensity of the impinging signal up to a level that is high enough to trigger the nonlinear

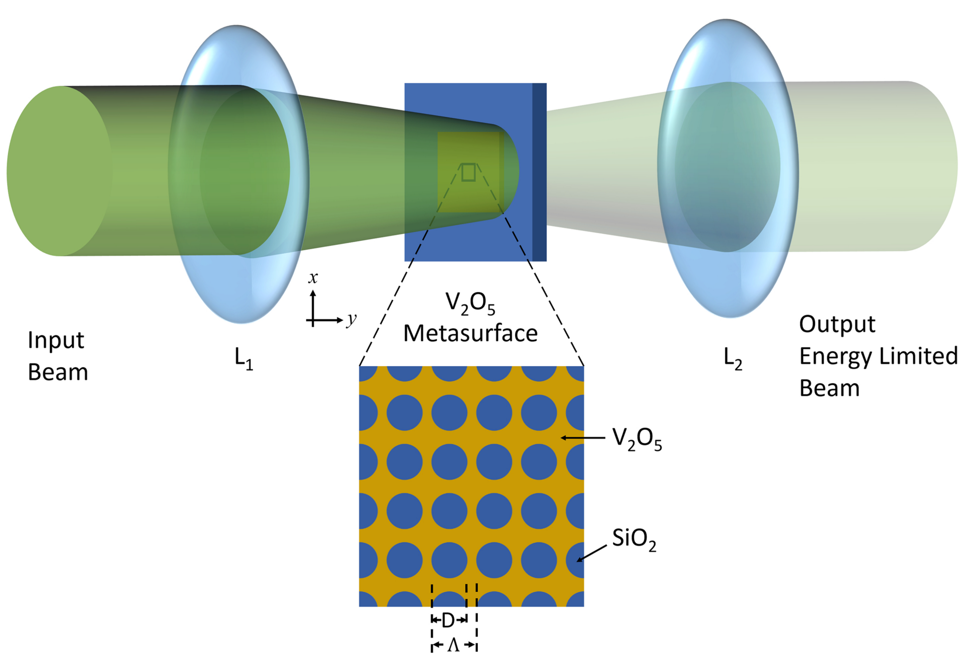

FIG. 1. A representative schematic of the proposed nonlinear optical metasurface consisting of a periodic array of air holes in a V_2O_5 layer, deposited on a silica substrate. The input laser beam becomes focused by the first lens to a high intensity spot, at the position of the V_2O_5 metasurface. The second lens collimates the light transmitted through the metasurface.

phenomena in the constituent crystal. Once the light passes through the metasurface, it is collimated by the second lens.

II. METHOD

We have developed an in-house FDTD solver to rigorously model the nonlinear refraction and absorption. The developed solver is based on the method outlined by Suzuki[16] for introducing the complex third-order nonlinear susceptibility $\left(\chi^{(3)}\right)$ in FDTD. V_2O_5 is modeled as an isotropic nonlinear medium. The polarization model that we use is as follows:[17]

$$\vec{P} = \vec{P}^{(1)} + \vec{P}^{(3)} = \varepsilon_0 \left[\chi^{(1)} + \chi^{(3)}\right] \vec{E} \tag{1}$$

Moreover,

$$\chi^{(3)} = \chi^{(3)}_{\text{Kerr}} + \chi^{(3)}_{\text{TPA}} \tag{2}$$

where, $\chi^{(3)}_{\text{Kerr}} = \chi^{(3)}_0 |\vec{E}|^2$ is the real-part of the third-order nonlinear susceptibility representing nonlinear refraction, and

$$\chi^{(3)}_{\text{TPA}} = i\frac{c_0 n_0 \beta_{\text{TPA}}}{\omega} I = i\frac{2c_0^2 \varepsilon_0 n_0^2 \beta_{\text{TPA}}}{\omega} |\vec{E}|^2 \tag{3}$$

is its imaginary part and responsible for nonlinear absorption. In eq.(3), c_0 is speed of light in vacuum, ε_0 is vacuum permittivity, n_0 is refractive index of nonlinear medium, and β_{TPA} is two-photon absorption (TPA) coefficient.[16] It is noteworthy that, the preceding formulation is based on the $I = 2\epsilon_0 c n_0 |E|^2$ convention for light intensity.

III. RESULTS AND DISCUSSION

In this study, we consider the incident laser wavelength of 532 nm, which corresponds to the second harmonic of a doubled Nd-YAG+ laser. The geometrical dimensions in Fig. 1 are optimized to achieve the highest starting transmission with largest nonlinear drop-off. As a result, the thickness of the V_2O_5 layer is chosen to be h=135 nm, periodicity Λ=320 nm, and hole diameter is D = 255 nm. The refractive indices of substrate and V_2O_5 are considered as n_{sub}=1.46 (SiO_2) and $n_{V_2O_5}$ = 2.5, respectively. Finally, nonlinear parameters of V_2O_5 are $\chi_0^{(3)} = 6\times10^{-16} m^2/V^2$, and $\beta_{\text{TPA}} = 3.5\times10^{-5}$cm/W (from ref. 14).

The linear transmittance spectrum of the V_2O_5 metasurface, in the visible range, is depicted in Fig. 2. The metasurface has a high transmittance, about 0.9, for low values of the incident light intensity (linear regime) at λ=532 nm. Additionally, the metasurface undergoes a resonance around

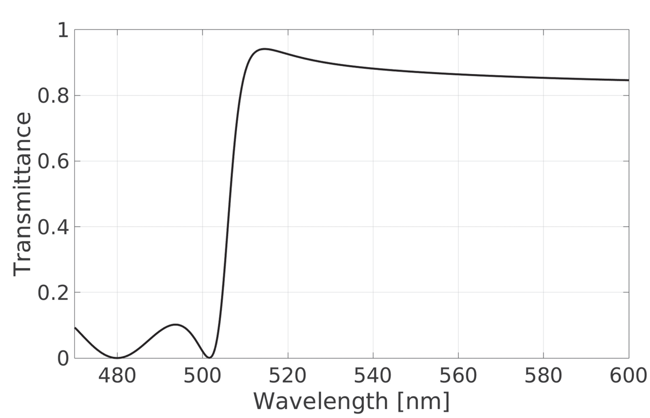

FIG. 2. Linear transmittance spectrum of the V_2O_5 metasurface of Fig. 1 in the visible range. The parameters of the metasurface along with optical properties of the employed materials are mentioned in the text. The metasurface has a hight transmittance at λ=532 nm, and undergoes a resonance around 505 nm.

505 nm, where the transmission drops and the surface becomes reflective. For this simulation, we assume no linear losses in the V_2O_5 material. Experimentally, the bandgap of V_2O_5 is found to be about 515 nm;[12,14,18] thus, our assumption of transparency is valid for the excitation wavelength of 532 nm.

As we show below (see Fig. 3), once the intensity of the incident irradiation increases, nonlinear losses cause the initial transmission of the metasurface at 532 nm to drop considerably. Moreover, the nonlinear refraction is responsible for a shift in the resonance frequency, which can further decrease the transmittance due to a redshift.[19] It is the aggregate effects of the aforementioned phenomena that lead to the large contrast between linear and nonlinear transmittances.

Next, we shall consider the numerically calculated transmittance, reflectance, and absorptance curves of the metasurface, versus the incident signal intensity, as depicted in Fig. 3 at the wavelength of 532 nm. Due to the nonlinear nature of the problem, the conventional broadband treatment in FDTD is not applicable here. Hence, we illuminate the metasurface with a normally incident plane wave with sinusoidal waveform. The FDTD update procedure is explained in detail in ref. 16. The transmission and reflection coefficients are calculated using the well known FDTD formulation for periodic structures.[20] Having the transmission and reflection coefficients, transmittance, T, and reflectance, R, can be readily obtained and subsequently, nonlinear absorptance, A, is given by $A = 1 - T - R$. We repeat the calculations for different values of the incident light intensity, represented by markers is Fig. 3.

The transmittance remains almost constant (≈0.92) for values of intensity lower than 10^6 W/cm^2. However, it drops off after this threshold and reaches 3.87×10^{-3} for I_{in} = 10^{10} W/cm^2. Hence, the output power reduces to $I_{out} = 3.87\times10^7$ by the metasurface. Meanwhile, the reflectance starts with a low value in the linear regime, and also decreases due to the nonlinear effects. Hence, the decrease of the transmittance can be fully attributed to nonlinear absorption. Output light intensity as a function of input intensity is also plotted in the inset. It can be observed that the output intensity saturates for values of input intensity larger than $I_{in} \approx 10^8$W/cm^2. It is worth mentioning that, V_2O_5 has previously demonstrated stability and reversible behavior under laser irradiation up to 10^9W/cm^2.[14]

Near-field plots of the x component of the electric field at λ=532 nm, for two extreme values of input light intensity in Fig. 3, namely, I_{in}=10^2W/cm^2 and I_{in}=10^{10}W/cm^2 are plotted in Figs. 4(a) and 4(b), respectively. The incident light is x-polarized, and the electric field is normalized to the amplitude of the incident signal. From Fig. 4(a), there is a strong field localization between two adjacent rows of the metasurface. As the incident light intensity increases, this field enhancement further boosts the nonlinear effects of the V_2O_5 layer; that is the main reason behind the large contrast between the values of low-intensity (linear) and high-intensity (nonlinear) transmittance, compared to an unpatterned film. There is a trade-off between optical limiting threshold and the transmittance contrast in designing the proposed metasurface. More specifically, for higher values of field enhancement, the optical limiting threshold will be lower (due to the boosted nonlinear absorption). However, higher field enhancement means working closer to the resonance wavelength

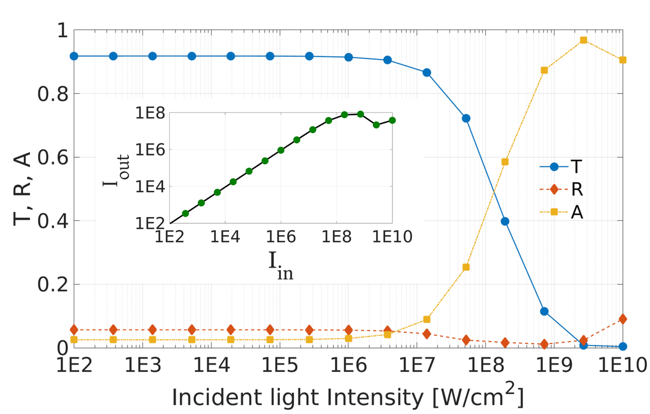

FIG. 3. Transmittance, reflectance, and absorptance curves of the metasurface of Fig. 1, versus the incident light intensity, at λ=532 nm. Output light intensity as a function of input intensity is also plotted in the inset.

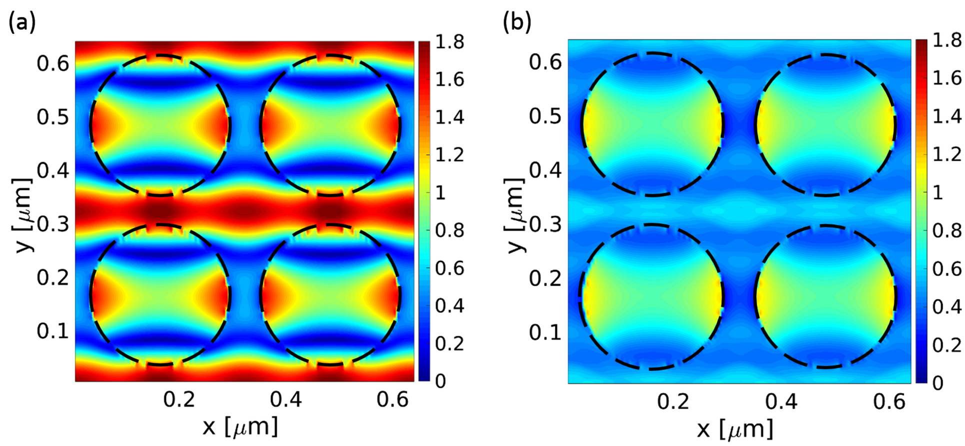

FIG. 4. Near-field plots of $|E_x|$ at λ=532 nm for, (a) $I_{in}=10^2$W/cm^2, and (b) $I_{in}=10^{10}$W/cm^2, in the middle of the V_2O_5 layer. The incident light is x-polarized, and the electric field is normalized to the amplitude of the incident signal.

(see Fig. 2), which generally leads to lower change in the transmittance due to the nonlinear effects. (see Fig. SpI-1 in the supplementary material for further illustration of this concept.)

From Fig. 4(b), the normalized field enhancement becomes smaller for large values of input intensity, compared to the linear regime. This phenomenon can be attributed to the increased nonlinear loss of the resonant structure (decreased quality factor), as well as, the change in the refractive index of V_2O_5 due to the Kerr effect, that leads to a shift in the resonance frequency. The change in the refractive index caused by the Kerr effect can be expressed as[16] $\Delta n = \chi_0^{(3)}|E|^2/2n_0$. Based on Fig. 4 (b), for $I_{in}=10^{10}$W/cm^2, the maximum electric field intensity inside the V_2O_5 layer is approximately 8.24×10^7V/m. Hence, maximum Δn can be estimated as 0.81.

To show that no temporal broadening occurs due to the nonlinear enhancement, we plot the transient response of the V_2O_5 metasurface to a modulated Gaussian incident waveform in the far-field region in Fig. 5. The standard deviation of the Gaussian pulse is chosen to be 40 fs (see Fig. SpI-2, SpI-3, and SpI-4 in the supplementary material for further transient responses.)

Finally, the optical transmittance of the proposed V_2O_5 metasurface as a function of the input light intensity, is compared in Fig. 6 with that of an unpatterned V_2O_5 film. To facilitate this comparison, we fit the transmittance response of the metasurface to the nonlinear Beer's law to estimate its "effective" two-photon coefficient. The V_2O_5 film has the same thickness as the metasurface, and

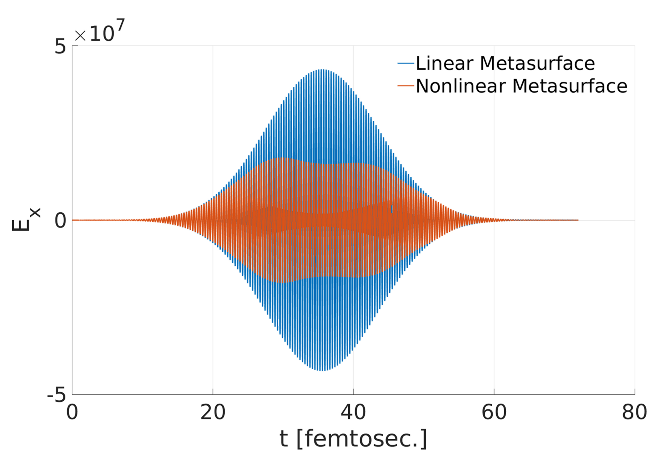

FIG. 5. Transient response of the V_2O_5 metasurface to a modulated Gaussian incident waveform in the far-field region. The peak intensity of the input signal is 10^9 W/cm^2. The response of the linear metasurface with the same refractive index as V_2O_5 is also plotted for reference.

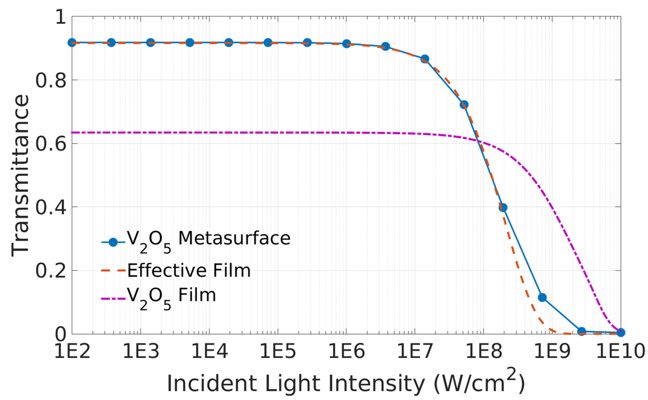

FIG. 6. Optical transmittance as a function of the incident light intensity at λ =532nm, for the proposed V_2O_5 metasurface, a V_2O_5 film with the same thickness and nonlinear absorption coefficient (h=135nm, β=3.5×10^{-5}cm/W), and an effective film, that shows a nonlinear response similar to the metasurface (h_{eff}=191nm, β_{eff}=27×10^{-5}cm/W).

the parameters of the effective film are: h_{eff}=191nm, and β_{eff}=27×10^{-5}cm/W. It can be observed that for the V_2O_5 film, the linear transmittance is 0.64 compared to 0.92 for the metasurface, which leads to a smaller nonlinear contrast. In order to compare the effectiveness of the metasurface to an unpatterened film, we compute an I_{50} parameter, an intensity value at which the transmittance drops to 50% of its initial, low intensity value.[21] For the patterned metasurface, I_{50} = 1.5×10^8W/cm^2, whereas for the nonlinear unpatterned film of equivalent thickness I_{50} = 2×10^9W/cm^2. Thus, an order of magnitude improvement in effective nonlinearity is obtained for a resonant metasurface. Additionally, the starting transmission of the resonant metasurface is 30% higher than the plain film due to the efficient suppression of reflection, thus obviating the need for an antireflecting coating layer, which itself might be prone to laser degradation.

IV. CONCLUSION

A low profile nonlinear metasurface comprising a periodic array of air holes in a V_2O_5 layer, aimed at protecting sensitive optical devices against high-power laser radiation is introduced for the first time in this letter and investigated numerically with a full-wave FDTD method. The proposed nonlinear metasurface increases the contrast between linear and nonlinear values of transmittance, and decreases the input intensity threshold for the nonlinear response, by more than an order of magnitude (compared to an unpatterned V_2O_5 film). While we choose V_2O_5 to illustrate the efficiency of our metasurface, the design is applicable to any material with a high initial transparency, high linear refractive index contrast and large intrinsic nonlinearity. Although the metasurface in this letter works in the transmission regime, it is totally feasible to achieve a metasurface in the reflection regime, just by employing the complimentary geometry, namely, an array of disks.

SUPPLEMENTARY MATERIAL

See supplementary material for a discussion on the the trade-off between the limiting threshold and transmittance contrast in designing the proposed nonlinear metasurface. Also, transient responses of the metasurface to a modulated step function and a modulated Gaussian pulse are presented in the supplementary material to investigate the response time and examine the performance of the device when illuminated by a pulsed laser, respectively.

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