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Optical Kerr nonlinearity of disordered all-dielectric resonant high index metasurfaces with negative refraction

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In recent years, nonlinear optical properties of all-dielectric high index metasurfaces have attracted a significant interest of researchers [1]. For example, silicon metasurfaces exhibited enhancement of third harmonic generation by several orders of magnitude compared to the massive material [2, 3]. The intensity-dependent refractive index is regularly used in designing all-optical compact switches. As demonstrated in [4], all-optical switching of femtosecond laser pulses passing through flat nanostructure of subwavelength silicon nanodisks at their magnetic dipolar resonance occurs owing to two-photon absorption being enhanced by a factor of 80 with respect to the unpatterned film. In [5], random monodisperse metasurfaces of gallium phosphide (GaP) spheres near Mie resonances were shown by three-dimensional finite-difference time-domain (FDTD) modeling to have an optical Kerr effect exceeding by two orders of intensity that of the bulk gallium phosphide. Moreover, being single negative metamaterials, the monodisperse metasurfaces with sphere sizes in the vicinity of the Mie resonances reveal the inversion of the sign of the second-order nonlinear refractive index [5]. The possibility of negative refraction by the disordered metasurface consisting of GaP spheres with two radii close to the first magnetic and electric Mie resonances was demonstrated in [6]. However, until now, the effective Kerr nonlinearity of the negative index metamaterials has not been evaluated.

In this work, the optical Kerr nonlinearity of random metasurfaces having the negative effective refractive index is investigated using three-dimensional FDTD simulations at the wavelength of 532 nm. The second-order nonlinear refractive index is also calculated for bidisperse mixtures with various sizes or concentrations of GaP spheres in proximity to the negative refraction regime.

The procedure of computation of the real part of the effective nonlinear refractive index of the monolayer nanocomposites is described in depth in [10]. The simulation parameters in the present study were same as given in [5]: the size of the FDTD computational domain was $4 \times 4 \times 30\,\mu$m, the space resolution of the simulations was 5 nm. The examined sample was a disordered monolayer comprising the equal numbers of GaP spheres of two radii surrounded by vacuum. As shown in [6], the densely packed bidisperse monolayer of GaP spheres with the radii r of 77 and 101 nm exhibits the negative refraction at $\lambda = 532$ nm.

The evaluation of the effective second-order nonlinear $n_{2\,\mathrm{eff}}$ refractive index for the bidisperse monolayer of the spheres with volume fraction $f = 25.7\,\%$, $r_1 = 77$ and $r_2 = 101$ nm having negative refraction index is $(6.5 \pm 0.8) \times 10^{-15}\,\mathrm{m}^2/\mathrm{W}$ which is two orders of magnitude larger than that of the bulk gallium phosphide. For purposes of comparison, the bidisperse monolayer consisting of spheres of artificial material with n_0 of GaP and n_2 of gallium phosphide with negative sign was modeled: the resulting value of $n_{2\,\mathrm{eff}} = -(6.5 \pm 0.7) \times 10^{-17}\,\mathrm{m}^2/\mathrm{W}$, that is the negative index metasurface does not show the inversion of $n_{2\,\mathrm{eff}}$ sign.

Then, the optical nonlinearity of the bidisperse metasurface during the concentration transition to the negative refraction state is investigated. The behavior of $n_{0\,\mathrm{eff}}$ through the transition to negative values was studied in [6]. Figure 1 illustrates the dependency of the effective second-order nonlinear refractive index $n_{2\,\mathrm{eff}}$ on the volume fraction of nanoparticles in the bidisperse monolayer. At low concentrations of the GaP spheres ($f = 8$–$18\,\%$), the metasurface exhibits the negative value of $n_{2\,\mathrm{eff}}$ due to the inversion of optical Kerr coefficient near the electric dipole Mie resonance [5]. For the higher volume fractions of nanoparticles, $n_{2\,\mathrm{eff}}$ becomes positive and has a peak when $n_{0\,\mathrm{eff}}$ crosses zero. Further, the effective second-order nonlinear refractive in-

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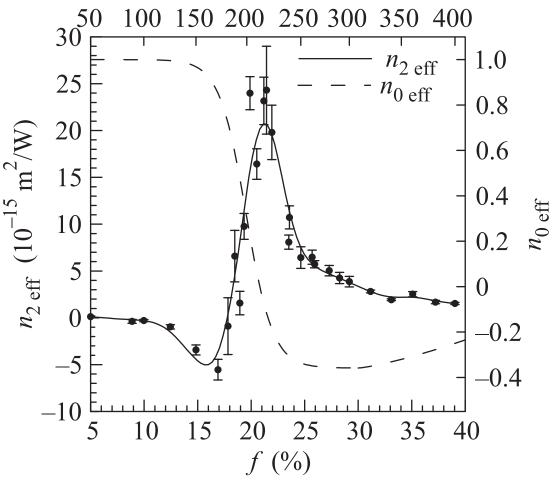

Fig. 1. Effective linear $n_{0\,\mathrm{eff}}$, second-order nonlinear $n_{2\,\mathrm{eff}}$ refractive indices of the disordered bidisperse metasurface consisting of equal numbers of GaP spheres with radii $r_1 = 77$ nm and $r_2 = 101$ nm as a function of volume concentration f. The upper abscissa axis displays the net number of the particles on $4 \times 4\,\mu$m area. The fit for effective linear refractive index $n_{0\,\mathrm{eff}}$ was taken from [6]. The error bars show the standard deviations

dex gradually decreases for the concentrations of spheres corresponding to the negative refraction. By this means, the disordered monolayer bidisperse metasurface has highest magnitudes of $n_{2\,\mathrm{eff}}$ under the conditions of zero index medium.

In conclusion, the nonlinear optical Kerr effect of bidisperse disordered monolayer nanocomposites of GaP spheres is studied numerically. It is displayed that this nanostructure possesses the maximum value of the second-order refractive index when the metasurface represents the mixture of nanoparticles with sizes just above the electric and magnetic dipole Mie resonances which results in the negative effective refractive index. The effective Kerr coefficient of the negative index all-dielectric metasurface has the same sign as that of the nanoparticle material. The bidisperse monolayer metasurfaces with different values of GaP sphere concentration exhibit a peak of the nonlinear optical Kerr effect when the effective linear index of refraction is close to zero. The monolayer metasurface shows the highest magnitudes of the second-order nonlinear refractive index as compared to that of thicker bidisperse nanocomposites.

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