Ultrathin gradient nonlinear metasurface with a giant nonlinear response: supplementary material

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Published 8 March 2016

This document provides supplementary material to “Ultrathin gradient nonlinear metasurface with a giant nonlinear response,” <http://dx.doi.org/10.1364/optica.3.000283>. © 2016 Optical Society of America

<http://dx.doi.org/10.1364/optica.3.000283.s001>

Efficient frequency conversion in ultrathin nonlinear metasurfaces

We start with the classical expression for a phase-matched second harmonic generation (SHG) in a nonlinear medium of thickness d in the plane-wave approximation [1,2]:

$$I_{2\omega} = \frac{(2\omega)^2}{8\epsilon_0 n_\omega n_{2\omega} c^3} |\chi_{\text{eff}}^{(2)}|^2 d^2 I_\omega^2, \quad (\text{S1})$$

where I_ω ($I_{2\omega}$) and n_ω ($n_{2\omega}$) are the intensity of the beam and the refractive index of the nonlinear material, respectively, at frequency ω (2ω), $\chi_{\text{eff}}^{(2)}$ is the complex value of the effective nonlinear susceptibility of the material for a particular input-output beams polarization [1,2], c is the speed of light in vacuum, and d is the materials thickness. The intensity of the pump electromagnetic wave can be expressed through the electric field amplitude as

$$I_\omega = 2n_\omega \epsilon_0 c |E_\omega|^2, \quad (\text{S2})$$

where we follow the formalism of Refs. [1,2] in which the real E-field in the electromagnetic wave is given as $\vec{E}(t) = \vec{E}_\omega e^{j\omega t} + c.c.$ Combining Eqs. (S1) and (S2) we obtain the expression for SHG conversion efficiency

$$\eta \equiv \frac{I_{2\omega}}{I_\omega} = \frac{\pi^2}{n_\omega n_{2\omega}} \left(\frac{d}{\lambda_{2\omega}} \right)^2 |\chi_{\text{eff}}^{(2)} E_\omega|^2 \approx \left(\frac{d}{\lambda_{2\omega}} \right)^2 |\chi_{\text{eff}}^{(2)} E_\omega|^2, \quad (\text{S3})$$

where E_ω is the pump electric field, $\lambda_{2\omega}$ is the SH wavelength, and we have used the undepleted pump approximation. Equation (S3) shows that, in order to provide high nonlinear conversion

efficiency in films of sub-wavelength thickness, we need to have $|\chi_{\text{eff}}^{(2)} E_\omega| \sim 1$ or, in other words, the nonlinear polarization $\vec{P}^{(2)} = \epsilon_0 \tilde{\chi}^{(2)} \vec{E}_\omega \vec{E}_\omega$ must become comparable to the linear one $\vec{P} = \epsilon_0 \tilde{\chi}^{(1)} \vec{E}_\omega$, where $\tilde{\chi}^{(1)} \sim 1$ is the linear susceptibility. Similar conclusions may be drawn for other three-wave mixing processes (sum- or difference-frequency generation), and for higher-order nonlinear processes.

Analytical model of spatial phase modulation

The SH response of the proposed nonlinear metasurface can be described using the effective local nonlinear susceptibility tensor $\chi_{\text{eff},ijk}^{(2)}$, and neglecting saturation effects, as [3]

$$\begin{aligned} \chi_{ijk}^{(2)} &= \frac{\chi_{MQW,zzz}^{(2)}}{V} \int_{UC} \frac{E_{z(i)}^{2\omega}(\mathbf{r})}{E_{inc(i)}^{2\omega}} \frac{E_{z(j)}^\omega(\mathbf{r})}{E_{inc(j)}^\omega} \frac{E_{z(k)}^\omega(\mathbf{r})}{E_{inc(k)}^\omega} dV \\ &= \chi_{MQW,zzz}^{(2)} \frac{\int_{UC} \xi_i^{2\omega}(\mathbf{r}) \xi_j^\omega(\mathbf{r}) \xi_k^\omega(\mathbf{r}) dV}{V} \\ &\equiv \chi_{MQW,zzz}^{(2)} I_{ijk}^{\text{overlap}}, \end{aligned} \quad (\text{S4})$$

where I_{ijk}^{overlap} (with $i, j, k = \{x, y\}$) denotes the modal overlap integral that provides the enhancement of the intrinsic $\chi_{MQW,zzz}^{(2)}$ of MQW, $E_{z(i)}^\omega$ ($E_{z(i)}^{2\omega}$) is the z-component of the electric field induced in the MQW at frequency ω (2ω) by an i -polarized impinging field $E_{inc(i)}^\omega$ ($E_{inc(i)}^{2\omega}$), and ξ_i^ω ($\xi_i^{2\omega}$) are the local enhancement factors,

defined as the ratio of the induced z -polarized E-field in the MQW to the E-field amplitude of i -polarized incident wave ω (2ω). This expression relates the average nonlinear polarization density in the metasurface to the impinging pump wave. Note that the overlap integral (S4) between the field distribution at fundamental and second harmonic frequency automatically accounts for the presence of any type of symmetry in unit cells. If the metasurface consists of identical unit cells, then under normal illumination by a plane wave the radiated field will be also a plane wave propagating normally away from the metasurface.

Let us consider now the scenario in which neighboring cells are weakly coupled, so that the effective local nonlinear susceptibility tensor $\chi_{ijk}^{(2)}$ in each of them is nearly independent from its neighbors. If the cells contain polarization-sensitive elements, like plasmonic resonators responding to particular field polarizations, then the nonlinear response will depend on their orientation. If we rotate all the elements in the metasurface by an angle φ around the z -axis (see Fig. 3 of the manuscript), the new effective nonlinear susceptibility of the metasurface $\chi_{ijk}^{(2)}(\varphi)$ will be equal to

$$\chi_{ijk}^{(2)}(\varphi) = \sum_{i'j'k'} R_{ii'}(\varphi) \chi_{i'j'k'}^{(2)} R_{jj'}(-\varphi) R_{kk'}(-\varphi), \quad (S5)$$

where $\chi_{ijk}^{(2)} \equiv \chi_{ijk}^{(2)}(\varphi=0)$ is the susceptibility tensor of a not rotated cell, and $R_{ij}(\varphi)$ is a standard rotation matrix

$$R_{ij}(\varphi) = \begin{pmatrix} \cos \varphi & -\sin \varphi \\ \sin \varphi & \cos \varphi \end{pmatrix}. \quad (S6)$$

We note that such metasurface with all rotated elements is still radiating normally. From Eqs. (S5) and (S6) it is seen that in the $\{x, y\}$ basis the dependence of tensor components on the elements' orientation φ is rather complicated. Let us rewrite Eqs. (S2) and (S3) in a circular polarization (CP) basis $\{R, L\}$ associated with right-hand and left-hand circularly polarized waves (RCP and LCP, respectively). For the rotation matrix we have

$$R_{\alpha\beta}^{\pm} = \sum_{ij} [\Lambda^{\pm}]_{\alpha i} R_{ij} [(\Lambda^{\pm})^{-1}]_{j\beta} = \begin{pmatrix} \exp(\mp i\varphi) & 0 \\ 0 & \exp(\pm i\varphi) \end{pmatrix}, \quad (S7)$$

where $\alpha, \beta = \{R, L\}$, and $\Lambda^{\pm}$ is a transformation matrix for CP waves propagating in $\pm z$ direction,

$$\Lambda_{\alpha i}^{\pm} = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & \mp i \\ 1 & \pm i \end{bmatrix}. \quad (S8)$$

For the susceptibility tensor we have

$$\chi_{\alpha\beta\gamma}^{(2)}(\varphi) = \sum_{\alpha'\beta'\gamma'} R_{\alpha\alpha'}^+(\varphi) \chi_{\alpha'\beta'\gamma'}^{(2)} R_{\beta\beta'}^-(\varphi) R_{\gamma\gamma'}^-(\varphi), \quad (S9)$$

with $\alpha, \beta, \gamma = \{R, L\}$, and

$$\chi_{\alpha\beta\gamma}^{(2)} \equiv \chi_{\alpha\beta\gamma}^{(2)}(\varphi=0) = \sum_{ijk} [\Lambda^+]_{\alpha i} \chi_{ijk}^{(2)} [(\Lambda^-)^{-1}]_{j\beta} [(\Lambda^-)^{-1}]_{k\gamma}. \quad (S10)$$

In Eqs. (S9) and (S10) we take into account that the pump and second harmonic waves propagate along $-z$ and $+z$ directions, respectively. Substituting (S7) into (S9) and using the fact that $R_{\alpha\beta}^{\pm}$ is diagonal, we obtain

$$\chi_{\alpha\beta\gamma}^{(2)}(\varphi) = R_{\alpha\alpha}^+(\varphi) \chi_{\alpha\beta\gamma}^{(2)} R_{\beta\beta}^-(\varphi) R_{\gamma\gamma}^-(\varphi). \quad (S11)$$

In particular, it is easy to show that

$$\begin{aligned} \chi_{RRR}^{(2)}(\varphi) &= \chi_{RRR}^{(2)} \exp(-i3\varphi), \\ \chi_{LRR}^{(2)}(\varphi) &= \chi_{LRR}^{(2)} \exp(-i\varphi), \\ \chi_{RLL}^{(2)}(\varphi) &= \chi_{RLL}^{(2)} \exp(+i\varphi), \\ \chi_{LLL}^{(2)}(\varphi) &= \chi_{LLL}^{(2)} \exp(+3i\varphi), \end{aligned} \quad (S12)$$

which are the expressions employed in the main text. From Eq. (S12) it is evident that under CP normal illumination of the metasurface with all elements rotated by an angle φ , the RCP and LCP components of the second harmonic signal will acquire different phase shift. We want to emphasize that Eqs. (S12) were derived for SHG in the "reflection" regime. Similar expressions can be easily obtained for a SHG metasurface operating in transmission. With the use of Eq. (S12), we can now design a metasurface able to tailor the SH RCP or LCP wave phase profile at will. If the unit cells are subwavelength and their orientations change adiabatically from element to element as $\varphi(x, y)$, the spatial variation of the local effective nonlinear susceptibility tensor will be

$$\begin{aligned} \chi_{RRR}^{(2)}(x, y) &= \chi_{RRR}^{(2)} \exp[-i3\varphi(x, y)], \\ \chi_{LRR}^{(2)}(x, y) &= \chi_{LRR}^{(2)} \exp[-i\varphi(x, y)], \\ \chi_{RLL}^{(2)}(x, y) &= \chi_{RLL}^{(2)} \exp[+i\varphi(x, y)], \\ \chi_{LLL}^{(2)}(x, y) &= \chi_{LLL}^{(2)} \exp[+3i\varphi(x, y)]. \end{aligned} \quad (S13)$$

Intersubband Absorption Measurements

Fig. S1(a) shows the experimentally-measured intersubband absorption spectrum after background correction. The measurement setup is shown in Fig. S1(b). Our measurement procedure is similar to that described in [3]. Assuming unpolarized mid-infrared (mid-IR) light illumination from the Fourier Transform Infrared Spectrometer (FTIR) thermal source, the intersubband absorption coefficient α_w is given as:

$$\alpha_w = -\frac{1}{L_{int}} \ln(10) \log_{10} \left(\frac{I_{TM}}{I_{TE}} \right), \quad (S14)$$

where L_{int} is the interaction length defined as

$$L_{int} = \frac{n_p L_{QW} N}{\cos(\theta)} \quad (S15)$$

θ is the angle of incidence with respect to the normal to the plane of the layers (45° in our case), L_{QW} is the thickness of each coupled quantum well section ($L_{QW}=15$ nm in our case), N is the total number of sections in the multi-quantum well (MQW) sample ($N=26$ in our case), and n_p is the number of passes through the MQW layer. Given our sample geometry (4.9 mm in length and 0.36 mm in thickness), we estimate $n_p=6$. From the data in Fig. S1a, we obtain transition $\hbar\omega_{21} \approx 150$ meV and $\hbar\omega_{31} \approx 248$ meV in good agreement with calculated values of 150 meV and 253 meV, respectively. The measured transition linewidths are $2\hbar\gamma_{12} \approx 24.8$ meV, $2\hbar\gamma_{13} \approx 29.2$ meV and the peak absorption $\alpha_w=1.01 \times 10^4$ cm $^{-1}$ and 3.09×10^3 cm $^{-1}$ for 1-2 transition and 1-3 transition, respectively. Given close agreement between calculated and measured transition energies and typically high accuracy of bandstructure calculations, we assume the transition dipole moments for intersubband transitions between states 1,2, and 3 in our structure are the same as simulated in Fig. 1a in the main text. The peak values of intersubband absorption for 1-2 and 1-3 transitions then allow us to estimate the actual doping level N_e in our structure, which may be different from the nominal doping

Fig. S1. (a) Intersubband absorption spectrum after background correction. (b) Intersubband absorption measurement setup. Unpolarized mid-IR light from FTIR passes through a linear polarizer and is focused onto one facet of MQW sample. Mid-IR light passed in multipass geometry of MQW sample is then detected from a photodetector.

used during growth. To that end, we express α_w through the dielectric constant for out-of-plane ($\epsilon_{\perp}$) E-field polarization as

$$\alpha_w = \frac{4\pi}{\lambda} \text{Im}(\sqrt{\epsilon_{\perp}}), \quad (\text{S16})$$

where

$$\epsilon_{\perp}(\omega) \approx \epsilon_{\text{core}}(\omega) + \frac{N_e (ez_{12})^2}{\epsilon_0 \hbar ((\omega_{21} - \omega) - i\gamma_{21})} + \frac{N_e (ez_{31})^2}{\epsilon_0 \hbar ((\omega_{31} - \omega) - i\gamma_{31})}. \quad (\text{S17})$$

and ϵ_{core} is the dielectric constant of the undoped heterostructures, ω is the pump frequency, e is the electron charge, N_e is the average bulk doping density, $\hbar\omega_{ij}$, $\hbar\gamma_{ij}$ and ez_{ij} are the energy, linewidth and dipole moment, respectively, for the transition between states i and j . Plugging the experimental values of $\hbar\omega_{ij}$ and $\hbar\gamma_{ij}$, and calculated transition dipole moments z_{ij} into the expressions above and assuming $N_e = 1.13 \times 10^{18} \text{ cm}^{-3}$ we obtain $\alpha_w \approx 1.01 \times 10^4 \text{ cm}^{-1}$ for 2-1 resonance and $\alpha_w \approx 3.09 \times 10^3 \text{ cm}^{-1}$ for 3-1 resonance, in close agreement with measurements. We use the value of $N_e = 1.13 \times 10^{18} \text{ cm}^{-3}$ for $\chi^{(2)}$ calculations. We note that this experimentally-deduced value of N_e is a factor of 1.3 smaller than the nominal doping density.

The MQW dielectric constant for the in-plane E-field polarization ($\epsilon_{\parallel}$) is calculated as

$$\epsilon_{\parallel}(\omega) \approx \epsilon_{\text{core}}(\omega) + i \frac{N_e e^2 \tau_D}{\epsilon_0 \omega m^* (1 - i\omega \tau_D)}, \quad (\text{S18})$$

where $\tau_D \approx 10^{-13} \text{ s}$ is a Drude relaxation time and we assumed free electron motion in the plane of semiconductor layers. The calculated dielectric constants of the MQW for the out-of-plane ($\epsilon_{\perp}$) and in-plane ($\epsilon_{\parallel}$) E-field polarization are used for computation of antenna resonances and E-field distribution in the MQW layer. A commercial Maxwell's equation solver based on the finite-integration method (CST studio) was used for computations. We note that quantum confinement due to etching is not expected to influence intersubband transition energies in our structures because the smallest feature size of our nanoresonators is 300 nm, much larger than the well width of 1-3 nm in our MQWs. Furthermore, because the electron dynamics of intersubband

transitions is determined by extremely fast (~ 0.3 -3 ps) longitudinal optical photon scattering, scattering due to surface defects is not expected to introduce significant modification to intersubband transition linewidths.

References

1. R. W. Boyd, *Nonlinear Optics* (Academic Press, 2008).
2. Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley-Interscience, 2002).
3. J. Lee, M. Tymchenko, C. Argyropoulos, P.-Y. Chen, F. Lu, F. Demmerle, G. Boehm, M.-C. Amann, A. Alù, and M. A. Belkin, "Giant nonlinear response from plasmonic metasurfaces coupled to intersubband transitions," *Nature* **511**, 65–69 (2014).