Orders of magnitude enhancement of optical nonlinearity in subwavelength metal-nonlinear dielectric gratings

Daniel J. Ironside and Jung-Tsung Shen
Department of Electrical and Systems Engineering, Washington University in St. Louis, St. Louis, Missouri 63130, USA

(Received 9 September 2012; accepted 2 January 2013; published online 16 January 2013)

We show that the optical properties of a metal-nonlinear dielectric grating can be precisely described by a uniform nonlinear dielectric slab. Based upon such a metamaterial mapping, we show that the effective optical nonlinearity in a metal-dielectric grating can be enhanced by orders of magnitude higher than that of the underlying nonlinear dielectric material, is broadband, and can be operated at a low quality-factor regime so as to have an extremely short intrinsic temporal response of a few picoseconds. Furthermore, we demonstrate extraordinary harmonic generation efficiency enhancement and large threshold-power reduction in bistability. © 2013 American Institute of Physics. [http://dx.doi.org/10.1063/1.4776697]

Materials with large optical nonlinearities and fast response and recovery times are essential for compact, ultrafast low-power nonlinear optical devices for switching and modulation, which are critical for all-optical information processing. Unfortunately, the intrinsic nonlinearities in naturally occurring materials are relatively weak, thus typically requiring huge operational powers, large interaction lengths, or both. The notion of enhancing nonlinearity by compressing the electrical energy into a very small volume, wherein weakly nonlinear materials are placed, has been used in the microwave community for sometime and has been readily adopted in optics research. Achieving enhanced optical nonlinearity at subwavelength scales offers exciting prospects for manipulating light, enabling miniaturization of nonlinear optical devices for integrated photonics. With the advances of nanofabrication techniques, subwavelength localization of the electric fields has been achieved in a variety of artificially created micro- and nanostructures. In the quest for enhanced optical nonlinearity, the subwavelength nonlinear optics has emerged as a unique and promising candidate, as it offers unprecedented opportunities for structural enhancement of nonlinear effects that can be modified by geometry.

Nonetheless, fundamental challenges exist in the implementations of nanostructured nonlinear optical components, despite the demonstrated superior performances. The modulation speed and the available fractional bandwidth of the configurations of high quality-factor (high-Q) nonlinear resonators are reduced by the large value of Q. In slow-light structures, the available bandwidth shrinks proportionally to the small group velocity \( v_g \). For slot waveguides, only part of the input light energy is used for enhancement, as the optical confinement \( \Gamma \) (the fraction of power confined and guided in the interested regions) is fundamentally limited (for example, \( \Gamma \approx 30\% \) in an optimal Si-SiO\(_2\) slab configuration with a 50-nm-wide slot).

In this letter, we show that an appropriately designed subwavelength metal-nonlinear dielectric (MNLD) grating system (Fig. 1(a)), consisting of alternating layers of metal and dielectric, can achieve extraordinary enhancement of nonlinearity and, at the same time, is capable of ultra-fast and broadband operation. It has been shown theoretically that a metal-linear dielectric grating can be optically transparent for TM-polarized light, with an effective refractive index entirely controlled by the characteristics of the grating. Such an artificial dielectric behavior of metallic grating has recently been demonstrated experimentally in the terahertz frequency range. At resonant frequencies, the transmission \( T \approx 1 \); the light squeezes and propagates through the subwavelength dielectric channels between metallic layers, producing significant electric field enhancement within the channels (Fig. 1(b)). Building on these achievements, we show that the MNLD grating can be precisely mapped into a homogenous nonlinear thin film. Such a metamaterial description provides deeper physical insights and quantitative criteria for gauging the enhancement of nonlinear processes in the MNLD grating. Moreover, the metamaterial approach also makes it possible to engineer both the effective linear and nonlinear optical properties of the grating by modifying the geometry. In particular, the MNLD grating can be designed to operate at a low-Q regime so that the cavity effect (round-trip times) is minimized to yield an extremely short intrinsic temporal response time approaching the single-pass fundamental limit. Our results address the central question regarding enhancing nonlinearity using the MNLD grating structures that has not been answered in previous investigations, namely, whether the prerequisite tight spatial electric field confinement, which only occurs in a fraction of the total volume of the grating, is sufficient to enhance the nonlinear processes. This is analogous to increase the traffic density by narrowing the road rather than slowing the traffic velocity.

By employing either energy considerations or a homogenization procedure, the MNLD grating can be mapped into a homogeneous nonlinear slab. The physical quantities of the two systems are related by \( \varepsilon(E) = \frac{d}{a} \varepsilon_0(E), \mu = \frac{s}{a}, \) and \( L = \frac{1}{\gamma} \). Here \( \varepsilon(E) \) is the field-dependent relative permittivity of the dielectric in the grating (the relative permeability \( \mu \) is assumed to be 1), \( d \) is the periodicity, \( a \) is the channel width, and \( L \) is the thickness (Fig. 1(a)). Parameters
associated with the dielectric slab are denoted with a bar. Choosing different $s$ allows the mapping to become purely dielectric ($s = d/a$, $\mu = 1$), purely magnetic ($s = d/a\epsilon$, $\bar{\epsilon} = 1$), or hybrid. For purely dielectric mapping, $\bar{\mu} = \mu = 1$, $L = \frac{L}{d/a}$, and $\bar{\epsilon}(\bar{E}) = (\frac{d}{a})^2 \epsilon(E)$. Equating the potential difference over one periodicity in the grating ($a \cdot E$) and in the mapped slab ($d \cdot \bar{E}$), the electric fields are related by $\bar{E} = \frac{a}{d} E$. When expressed in terms of the electric field $\bar{E}$ in the mapped slab, the second-order nonlinearity scales as $\bar{\epsilon} = (\frac{d}{a})^2 \epsilon + (\frac{d}{a})^3 \chi^{(2)} \bar{E} \equiv \bar{\epsilon}_l + \bar{\chi}^{(2)} \bar{E}$ and the third-order nonlinearity scales as $\bar{\epsilon} = (\frac{d}{a})^2 \epsilon + (\frac{d}{a})^3 \chi^{(3)} |\bar{E}|^2 \equiv \bar{\epsilon}_l + \bar{\chi}^{(3)} |\bar{E}|^2$. $\epsilon_l$ is the linear permittivity. Thus, $\bar{\epsilon}_l = (\frac{d}{a})^2 \epsilon_l$, $\bar{\chi}^{(2)} = (\frac{d}{a})^4 \chi^{(2)}$, and $\bar{\chi}^{(3)} = (\frac{d}{a})^6 \chi^{(3)}$. For $n$th-order susceptibility, $\bar{\chi}^{(n)} = (\frac{d}{a})^{n+1} \chi^{(n)}$. The enhancement is purely geometrical and does not require phase-matching condition. Note that $\chi^{(2)}$ and $\chi^{(3)}$ in general are both tensor with components relating to the symmetry of the nonlinear materials. As the MNLD gratings only support propagating modes with an electric field along the grating direction $n$ (Fig. 1(a)), the electric field of the incoming light $E_0$ and the generated non-linear polarizations $P_{2\omega}$ and $P_{3\omega}$ (hence the harmonic fields) must have non-vanishing component along the grating direction: $P_{2\omega} \cdot n = \sum_{jk} \left[ \sum_{l} \bar{\chi}^{(2)}_{jk}(n_l \cdot n) E \cdot \hat{n}_l \right] n_j (E \cdot \hat{n}_i) n_k$, where the unit vector $\hat{n} = (n_1, n_2, n_3)$. $\chi^{(2)}$ in the above scaling relations is the effective susceptibility. When $E$ is parallel to $\hat{n}$ (TM wave), $P_{2\omega} \cdot \hat{n}$ is nonzero if $\sum_{jk} \bar{\chi}^{(2)}_{jk} n_j n_k$ is nonzero. Similar expression holds for $\chi^{(3)}$.

The above simple mapping is only asymptotically accurate in the deep subwavelength regime ($\lambda/d \gg 1$ and $d/a \gg 1$). By introducing a single correction factor $\epsilon_c$ in the mapped linear permittivity so that $\bar{\epsilon}_l = \epsilon_c (\frac{d}{a})^2 \epsilon_l$, the validity of the mapping can be extended to the entire wavelength range of interest. $\epsilon_c$ is determined by equating the transmission spectra of both systems, $T'_\text{mapped}(\lambda) = T'_\text{grating}(\lambda)$, where $T'_\text{mapped}(\lambda)$ is the transmission spectrum of the dielectric thin film with the refractive index $n' = (d/a) \sqrt{\epsilon_c \epsilon_l}$ and thickness $L = L/(d/a)$, and $T'_\text{grating}(\lambda)$ is the transmission of the grating. Numerically $\epsilon_c$ is found to remain constant over the entire transmission peak. As the modification of the mapped linear permittivity slightly alters the fields within the slab, the nonlinear susceptibilities are slightly altered accordingly: $\bar{\chi}^{(2)} = \chi^{(2)} (\frac{d}{a})^4 \chi^{(2)}$ and $\bar{\chi}^{(3)} = \chi^{(3)} (\frac{d}{a})^6 \chi^{(3)}$, where $\chi^{(2)} = \left( \frac{|E|}{|d/a|} \right)^3$ and $\chi^{(3)} = \left( \frac{|E|}{|d/a|} \right)^4$, with $\bar{E}'$ being the corrected electric field in the thin film. These corrections are in general very small throughout the entire frequency range. For the grating system considered here, numerically $\chi^{(2)}_c$ and $\chi^{(3)}_c$ are essential ones. There is no free parameter in the mapping. The scaled mapping provides a highly accurate and predictive model for investigating the nonlinear optical responses of MNLD grating, requiring no parameter retrieval from numerical simulations or experimental measurements and, moreover, offers great computational flexibility; the nonlinear optical responses can be computed much more efficiently using a homogenous nonlinear dielectric slab. Such a computational advantage is independent of the underlying computational schemes used to compute the optical properties.

To verify the above theory, we numerically simulated the optical nonlinear processes by solving Maxwell’s equations using a finite-difference time-domain (FDTD) method. Our first validation is the enhancement of second (SHG) and third (THG) harmonic generation efficiency of the grating for normally incident TM-polarized light (Fig. 2). For SHG, the MNLD grating has the following parameters: $a = 0.165 \mu m$, $d = 1.325 \mu m$ ($d/a = 8$), and $L = 11.2 \mu m$, which was designed to be near resonance at the operating wavelength of $10.6 \mu m$ (CO2 line at mid-infrared rage) for optimal output. The $Q$-factor is 180. The second-order nonlinear material copper chloride I (cuprous chloride, CuCl) has $\epsilon_l = 3.5834$ and $\chi^{(2)} = 13.4 \times 10^{-12} m/V$. The scaled mapping predicts an enhanced linear permittivity $\epsilon_l = \epsilon_c (\frac{d}{a})^2 \epsilon_l = 231.46$ ($\epsilon_c = 1.009242$) and an enhanced second-order susceptibility $\chi^{(2)} = (\frac{d}{a})^4 \chi^{(2)} = 6860.8 \times 10^{-12} m/V$ for a uniform slab with thickness $L = 1.4 \mu m$. Fig. 2(a) plots the FDTD-calculated efficiency $I_{2\omega}/I_\omega$ versus the normalized input field strength $\sqrt{\chi^{(2)}} |E_0|$ ($I_\omega$ is the intensity of the input light and $E_0$ the field strength), which clearly shows an excellent agreement between the efficiencies of the MNLD grating and the mapped slab. To demonstrate the enhancement, the efficiencies of two uniform reference slabs of CuCl, one with a thickness of the grating $L$ (transmission $T = 1$) and one with a thickness of the scaled mapping $L$ (transmission $T = 0.6823$) are also plotted. Throughout the entire input field range plotted, the efficiency of the grating is at least two orders of magnitude larger than that of any reference slab with thickness between $L$ and $L$. Moreover,
the numerical results agree with the coupled-mode theory. In the undepleted pump regime, $I_{2\omega} \propto Q_{2\omega}^2 Q_{2\omega}^2 (\chi^{(3)} L)^2 T_0^2 / n_{2\omega}^2$ for homogeneous slab, which predicts an SHG enhancement $I_{2\omega} / I_{2\omega} > 1$ (where $Q_{2\omega}$, $Q_{2\omega}$, $n_{2\omega}$, $n_{2\omega}$ are the quality-factor and refractive index of the fundamental and harmonic frequency, respectively). In the high-Q limit, $Q \propto n = \omega / \alpha$, and the expression simplifies to $I_{2\omega} \propto (\chi^{(3)} L)^2 T_0^2$.

Fig. 2(b) shows the FDTD-calculated THG efficiencies. The MNLD grating has the following parameters: $a = 0.294 \mu m$, $d = 1.767 \mu m$ ($d/a = 6$), and $L = 2.648 \mu m$, with germanium (Ge) as the third-order nonlinear material ($\epsilon_l = 16.022$ and $\chi^{(3)} = 4.21 \times 10^{-18} m^2/V^2$ at 10.6 $\mu m$) and a $Q$-factor of 38. The scaled mapping predicts an enhanced linear permittivity $\tilde{\epsilon}_l = \epsilon_l (\frac{d}{a})^2 \epsilon_l = 584.30$ with $\epsilon_l = 1.013024$ and an enhanced third-order susceptibility $\tilde{\chi}^{(3)} = \left( \frac{d}{a} \right)^2 \chi^{(3)} = 5.46 \times 10^{-15} m^2/V^2$ for a slab of thickness $L = 0.441 \mu m$. The numerical simulations demonstrate an excellent agreement between the efficiencies of the grating and the scaled mapping. Furthermore, throughout the entire input field range plotted, the efficiency of the grating is at least three orders of magnitude larger than that of the reference Ge slab with thickness between $L$ (transmission $T = 0.275$) and $L$ ($T = 1$). The numerical results are also consistent with the predictions of the coupled-mode theory.

Our second validation shows that bistable switching can be achieved in a thin MNLD grating that is not possible by using the constituent nonlinear material only. The MNLD grating was chosen to have the following parameters: $a = 0.221 \mu m$, $d = 0.883 \mu m$ ($d/a = 4$), and $L = 4.24 \mu m$, with silicon (Si) as the third-order nonlinear material, operating at wavelength 10.6 $\mu m$ that is much larger than the grating thickness. At 10.6 $\mu m$, Si has a linear permittivity $\epsilon_l = 12.09$ and a third-order susceptibility $\chi^{(3)} = 2.63 \times 10^{-19} m^2/V^2$. The scaled mapping predicts an enhanced linear permittivity $\tilde{\epsilon}_l = \epsilon_l (\frac{d}{a})^2 \epsilon_l = 194.75$ with $\epsilon_l = 1.006445$ and an enhanced third-order susceptibility $\tilde{\chi}^{(3)} = \left( \frac{d}{a} \right)^2 \chi^{(3)} = 6.74 \times 10^{-17} m^2/V^2$ for a slab thickness of $L = 1.06 \mu m$. Fig. 3 shows that the bistability behaviors of both grating and mapped systems are essentially identical throughout the entire input strength range plotted. Moreover, the agreements for both low and high transmisons of the bistability window indicate that the scaled mapping is valid over the entire transmission peak. Fig. 3 also plots the bistability behavior of a uniform reference slab of Si with a much larger thickness 10.512 $\mu m \approx \lambda$, which exhibits the first on-set of bistability (it is numerically checked that no thinner Si reference slab exhibits bistability in the input range). The onset of the power threshold for MNLD grating starts at a much lower normalized input 0.026, which is about 50 times smaller than the onset power threshold for the reference slab at the normalized input at 1.26. The bistability window of the grating also exhibits larger contrast (intensity difference between the high and low output states). Moreover, from the transmission spectrum, the quality-factor value for the grating at the resonance is $Q = \lambda_{res} / \Delta \lambda_{HWHM} = 380$, where $\lambda_{res} = 10.6 \mu m$ and $\lambda_{HWHM} = 27.89 \mu m$ is the full-width-at-half-maximum resonance bandwidth. This corresponds to a cavity photon lifetime of $\tau_{cav} = \lambda_{res}^2 (2\pi) \Delta \lambda_{HWHM} = 2.1 \mu s$.
where $c$ is the speed of light in vacuum.\textsuperscript{27} We also observed from numerical simulation that the bistability can be switched between high-low states within ~30 optical cycle, which corresponds to 1 ps switching time. Thus, by operating at low-$Q$ regime, the temporal response of this thin MNLD grating can theoretically be as short as a few picoseconds. In contrast, the nonlinearity in low-$Q$ Fabry-Perot resonator is negligible due to its significantly low intra-cavity field intensity.\textsuperscript{28}

To demonstrate the underlying physics, we have so far assumed the metal to be perfect electric conductor (PEC), allowing no field penetration. In reality, the light signals dissipate as they propagate through the slits between metal layers. Here we show that orders of magnitude enhancement of optical nonlinearity can still be achieved in lossy metallic grating. We consider the SHG efficiency for silver grating with $a = 1 \, \mu m$, periodicity $d = 4 \, \mu m$ ($d/a = 4$), and thickness $L = 16.8 \, \mu m$, containing cadmium selenide (CdSe) nonlinear material. At the operating wavelength 10.6 $\mu m$, silver has a complex permittivity $\varepsilon_{Ag} = -4551.87 + i1446.23$,\textsuperscript{29} and CdSe has a linear permittivity $\varepsilon_i = 6.0614$ and a second-order susceptibility $\chi^{(2)} = 109 \times 10^{-12} \, m/V$.\textsuperscript{26} The grating was designed so that when assuming PEC boundary conditions, the transmission at the operating frequency is near resonance, with a quality-factor $Q$ equal to 61. Fig. 4 plots the SHG efficiency of the system and two uniform CdSe reference slabs with thickness $L_{res} = 17.2 \, \mu m$ ($T = 1$) and $L = 4.2 \, \mu m$ ($T = 0.98$), respectively. Here $L_{res} = 17.2 \, \mu m$ is the thickness that is most closest to the thickness of the grating $L = 16.8 \, \mu m$ and has a perfect transmission (thus the optimal efficiency). Orders of magnitude enhancement are evident for the grating designed. Efficiency enhancement is 14.3 compared to the uniform slab with thickness $L_{res}$, and 288 compared to the uniform slab with thickness $L$. The plotted normalized input field range $10^{-5} - 1.5 \times 10^{-4}$ is equivalent to an intensity range from $4.1 \times 10^{-4} - 9.2 \times 10^{6} \, W/cm^2$ ($4.1 \times 10^{-1} - 9.2 \times 10^6 \, mW/\mu m^2$), which is below the optical damage threshold even with the field enhancement in the slits. We note that the efficiency could be further optimized by designing the silver grating to operate at local maximum transmission peak with the plasmonic effects (here the transmission for the dissipative silver grating reduces to 0.55 and is pushed away from resonance due to the plasmonic effects).

Although only the refractive nonlinearity is demonstrated, the amplification mechanism using tight spatially confined electric field applies as well to absorptive nonlinearity. Finally, we note that in this letter we consider the SHG and THG from the MNLD grating at normal incidence for TM light. As the intensity of the transmitted harmonic waves oscillates as a function of the incidence angle (Maker fringes\textsuperscript{30,31}), usually there exists an optimum angle of incidence for the highest signal output. We have numerically checked the validity of the scaling relations for small incidence angles, and it remains to be an interesting question whether the MNLD grating can be mapped to the same homogeneous nonlinear slab for all incidence angles.

The authors acknowledge informative discussions with L. Wang and W. Pickard. The computations were performed at the Rosen Center for Advanced Computing at Purdue University through the support of the National Science Foundation XSEDE program.
