Nonlinear response of GaAs gratings in the extraordinary transmission regime

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We theoretically describe a way to enhance harmonic generation from subwavelength slits milled on semiconductor substrates in strongly absorptive regimes. The metal-like response typical of semiconductors, like GaAs and GaP, triggers enhanced transmission and nonlinear optical phenomena in the deep UV range. We numerically study correlations between linear and nonlinear responses and their intricacies in infinite arrays, and highlight differences between nonlinear surface and magnetic sources, and intrinsic \( \chi^{(2)} \) and \( \chi^{(3)} \) contributions to harmonic generation. The results show promising efficiencies at wavelengths below 120 nm, and reveal coupling of TE and TM polarizations for pump and harmonic signals. A downconversion process that can regenerate pump photons with polarization orthogonal to the incident pump is also discussed. © 2011 Optical Society of America

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Extraordinary transmission (ET) [1] has been studied across the electromagnetic spectrum in an attempt to clarify the dynamics of the process [2–6]. ET through periodic slit arrays depends on geometrical parameters, such as aperture size, thickness, and periodicity [7,8]. The negative real part of the permittivity of metals [9] allows the formation of surface modes that couple the light into the apertures, pushing it through the holes. However, all materials display a negative permittivity below the plasma frequency, but, traditionally, only metals have been considered good candidates for ET-related processes. III–V semiconductors have been ignored not because the real part of the permittivity becomes negative just below 270 nm [10], but because absorption is too high.

ET was recently demonstrated for a GaAs layer having a finite number of apertures at UV wavelengths, despite high absorption [11]. Here we show that, under extreme absorption conditions, a GaAs grating allows the formation of forbidden bands, similar to plasmonic bandgaps [12], that minimize absorption. At the same time, this dynamics leads to the enhancement of harmonic generation at UV wavelengths. Several studies performed on metal substrates and gratings have shown that cavity effects relying on the formation of surface plasmons are essential in nonlinear processes [7,13,14]. Unlike metals, harmonic generation in semiconductors is driven mostly by \( \chi^{(2)} \) contributions, while symmetry breaking at the surface and the action of the Lorentz force on bound electrons play a nontrivial role in cross coupling orthogonal polarization states [13,14]. In what follows, we demonstrate that harmonic components undergo bandgap effects related to the pump wavelength and its harmonics, and how phase locking [15–17] allows the generation, propagation, and transmission of light at wavelengths that are below cutoff for the subwavelength guide even under high absorption conditions [18]. Additionally, we report a downconversion process that regenerates pump photons having polarization orthogonal compared to that of the incident pump.

We begin our investigation by numerically studying the linear response of an infinitely periodic GaAs [10] grating. The layer has thickness \( w \) and slits of width \( a \) spaced with periodicity \( p \) [Fig. 1(a)]. The results were obtained using the finite element method (Comsol Multiphysics [19]), and by means of a time-domain fast Fourier transform beam propagation method [20] that was also used to obtain the nonlinear results. A scan of the linear properties in the frequency domain under TM polarization was performed for a structure having \( a = 32 \) nm, \( w = 60 \) nm, and \( p = 100 \) nm. Figure 1(b) shows that, in this case, a sufficiently small pitch avoids the formation of surface waves that typically occur at the onset of new diffraction orders and cavity modes. The transmittance [red curve, Fig. 1(b)] shows resonant behavior with a prominent feature near \( \lambda = 240 \) nm. This peak is associated with the formation of the fundamental TEM-like mode in the GaAs/Air/GaAs truncated waveguide, or slit, whose spectral position is governed by the parameters \( w \) and \( a \). The choice of \( w = 60 \) nm for this structure is not accidental. This combination of parameters places the Fabry–Perot fundamental mode precisely where the absorption of the semiconductor is maximum, proving that (i) the excitation of the TEM-like guided mode does not depend on the imaginary part of the permittivity, and that (ii) ET may be achieved in spite of the large absorption that characterizes semiconductors in the UV range [11]. At first sight, a transmittance of 45% at \( \lambda = 240 \) nm may not seem impressive [red curve, Fig. 1(b)]. However, one should compare this value with a transmittance of 0.0008% obtained for a uniform GaAs layer of the same thickness, also at \( \lambda \sim 240 \) nm.

Next we investigated the linear properties of an infinite GaAs grating having \( a = 32 \) nm, \( w = 60 \) nm, and variable pitch size, for both TM- and TE-polarized incident plane waves tuned at \( \lambda = 240 \) nm. As one may infer from Fig. 2, transmittance and reflectance for the two polarizations are quite different. TM-polarized light is transmitted through the grating and is characterized by the formation of a polaritonic bandgap, a condition that takes place for those surface polariton wavelengths that match...
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generation may be achieved by introducing structure are strong but different compared to metal gratings that are either empty or filled with semiconductor materials. In metal gratings, forward and backward generation closely follows the linear transmission profile, showing maximum efficiency whenever the transmission displays a resonance, becoming a minimum whenever forbidden states occur for the pump field. Resonant semiconductor gratings are different in several respects. For example, $\chi^{(2)}$ and $\chi^{(3)}$ contributions arise directly from solid portions of the grating, and not from the slit. Second, absorption now plays a more central role, as the ratio of real to imaginary parts of the dielectric constant is much different than for metals. Tuning the pump at 240 nm means that the SH field is tuned at 120 nm and TH falls at 80 nm. Varying pitch size (Fig. 4) reveals that the nonlinear response is influenced significantly by the linear properties in a different way: the forward generated harmonics experience the same forbidden states of the incident pump field. These states occur when pump wavelength matches the effective $\lambda_{app}$ associated with every interface of the grating. In contrast, the reflected harmonics are generated more efficiently inside the polaritonic bandgap, where a

anisotropic tensor. For a material like GaAs, the nonlinear Cartesian components may be written as $P^{NL}_i = \chi^{(3)}(E_i^2 E_j^2 + E_k^2) E_i$ [14], where $i$, $j$, and $k$ are Cartesian coordinates. We choose $\chi^{(3)} = 10^{-18}$ (m/V)$^2$, a value consistent with what is reported in the literature for somewhat longer wavelengths [22,23]. Another way to elicit a nonlinear response from GaAs is to include terms arising from symmetry breaking and the magnetic Lorentz force [13,14], both of which can modify the qualitative and quantitative aspects of harmonic generation. For example, in materials that have a $\chi^{(2)}$ tensor similar to that of GaAs, the introduction of surface and/or magnetic terms opens a new interaction channel that transforms TM-polarized pump photons into TE-polarized pump photons via sequential upconversion and downconversion processes that rotate the polarization of light [14]. Therefore, combining surface, magnetic, and volume nonlinearities favors the generation of harmonics of both polarizations [14]. As Fig. 3 shows, a TM-polarized pump [Fig. 3(a)] generates TM- and TE-polarized second harmonic (SH) and third harmonic (TH), as well as a TE-polarized pump signal, an effect that could pilot the development of a nonlinear UV polarizer (see Fig. 3).

The links between linear and nonlinear dynamics in this structure are strong but different compared to metal gratings that are either empty or filled with semiconductor materials. In metal gratings, forward and backward generation closely follows the linear transmission profile, showing maximum efficiency whenever the transmission displays a resonance, becoming a minimum whenever forbidden states occur for the pump field. Resonant semiconductor gratings are different in several respects. For example, $\chi^{(2)}$ and $\chi^{(3)}$ contributions arise directly from solid portions of the grating, and not from the slit. Second, absorption now plays a more central role, as the ratio of real to imaginary parts of the dielectric constant is much different than for metals. Tuning the pump at 240 nm means that the SH field is tuned at 120 nm and TH falls at 80 nm. Varying pitch size (Fig. 4) reveals that the nonlinear response is influenced significantly by the linear properties in a different way: the forward generated harmonics experience the same forbidden states of the incident pump field. These states occur when pump wavelength matches the effective $\lambda_{app}$ associated with every interface of the grating. In contrast, the reflected harmonics are generated more efficiently inside the polaritonic bandgap, where a

Fig. 1. (Color online) (a) Detail of an infinite array of slits 32 nm wide on a GaAs layer 60 nm thick; $p = 100$ nm. (b) Transmission function (red curve) and permittivity values of GaAs (blue and black curves).

Fig. 2. (Color online) Transmittance and reflectance for the GaAs grating; $\lambda = 240$ nm, $a = 32$ nm, $w = 60$ nm, and pitch size varies. TM (transmission, blue dashed curve; reflection, black curve with triangles) and TE (transmission, red curve with squares; reflection, dark blue crosses) polarizations were monitored. A polaritonic bandgap is centered at $\lambda_{app} = p$. The TE-polarized field is suppressed.

Fig. 3. (Color online) Pump and harmonic field intensities inside and near the nanocavities. The magnetic field intensities are depicted for (a) pump, TM-polarized (b) SH and (c) TH; TE-polarized (d) pump, (e) SH, and (f) TH.
virtually zero transmission is associated with maximum reflection and minimal absorption. For comparison, we report harmonic generation from a uniform, 60-nm-thick GaAs layer (dark blue solid lines in Fig. 4). This baseline value can be used to appreciate the enhancement of each harmonic signal when variable periodicity is considered. Figure 4 show that SH and TH fields experience additional forbidden states because the harmonics operate at twice and three times the frequency of the pump: regardless of pump behavior, the harmonics are constrained by the size of the wavelength relative to grating periodicity. For example, the TM-polarized SH is inhibited beginning when $p$ matches the unperturbed Air/GaAs polariton wavelength of the pump and SH. The same phenomenon is also evident for third-harmonic generation (THG) with appropriate pitch values.

We note that the TE-polarized SH signal arises from $\chi^{(2)}$, while the TM-polarized SH signal comes from symmetry breaking and the Lorentz force acting on bound electrons [14]. It is notable that the TE-polarized signals are generated and transmitted in a regime where all TE-polarized fields should be forbidden, regardless of periodicity (see Fig. 2). This result is due to phase locking between the pump and its harmonics, which helps to overcome both absorption and cutoff effects. Finally, we remark that the harmonics are generated far from any set coherence length (grating thickness is only 60 nm), and that the harmonics are tuned to 120 and 80 nm, wavelength ranges that are quite difficult to manage.

In conclusion, we have demonstrated the prospect of second-harmonic generation and THG at extreme UV wavelengths originating from $\chi^{(2)}$, $\chi^{(3)}$, symmetry breaking, and the magnetic Lorentz force, under conditions of ET in GaAs gratings. For certain geometrical parameters and excitation wavelengths, the total linear transmittance reaches 50%, thanks to the coupling of surface waves inside subwavelength-sized slits. Phase locking binds the harmonics to the pump field, and creates conditions that allow SH and TH fields to resonate on the grating, despite the large resonant absorption and waveguide cutoff conditions for TE-polarized fields. We have shown that it is possible to trigger a downconversion process that regenerates pump photons having polarization orthogonal compared to the incident pump field.

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