MODELLING AND OPTIMIZATION OF THE SECOND-HARMONIC RADIATION PATTERN IN DIELECTRIC NANOANTENNAS

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KEYWORDS
Second Harmonic Generation, dielectric nanoantennas, radiation pattern.

ABSTRACT

We present numerical results that describe how to engineer the radiation pattern of the second harmonic (SH) signal generated by AlGaAs on aluminum oxide all-dielectric nanoantennas. The SH beam divergence is minimized by coherent forward and backward scattering of the radiation emitted at grazing angles from the optical antenna toward a concentric grating grown on the aluminum oxide substrate, whereas the symmetry of the SH mode is converted by introducing a suitably-designed phase shift. The parameters of the structure are optimized through extensive numerical simulations and design guidelines for fabrication are provided.

INRODUCTION

Optical antennas are a promising research area for their potential application in various areas of nanotechnology: as a matter of fact, their ability to convert propagating radiation into localized subwavelength modes at the nanoscale (Taminiau et al. 2008; Novotny 2008; Koenderink 2009; Devilez et al. 2010; Novotny and van Hulst 2010; Dorfmuller et al. 2011; Miroshnichenko et al. 2011) makes optical aerials highly desirable in many different fields. Antennas have been in common use at radiofrequencies (RF) for more than a century, in a wide variety of applications; as a consequence, well-assessed design rules have been developed in time, and are now available, at RF, for molding the electromagnetic radiation (Balanis 1982). Recent advancements in the fabrication of devices at the nanoscale has allowed to bring many of the concepts of the RF aerials to optics, leading to the development of optical antennas consisting of properly engineered subwavelength metallic and/or dielectric structures (Novotny and van Hulst 2010). In recent years, semiconductor nanoparticles have emerged as a promising alternative to metallic ones for a wide range of nanophotonic applications based on localized resonant modes in the entire visible and near-IR spectral ranges (Ginn et al. 2012; Albella et al. 2014; Shcherbakov et al. 2014). Different particle geometries can be considered, but mainly spherical and cylindrical antennas have been considered to date. Nanodisks have proven versatile in tailoring of electric and magnetic response, by exploiting two degrees of freedom: radius and height (van de Groep and Polman 2013; Staude et al. 2013). All-dielectric optical antennas also offer unique opportunities for nonlinear optics at the nanoscale with two prominent assets that lack in plasmonics: very low losses leading to high radiation efficiency and multipolar characteristics of both electric and magnetic resonant optical modes potentially leading to the engineering of the radiation pattern (Shcherbakov et al. 2014). It is also worth saying that while in plasmonic nanostructures the optical nonlinear response is dominated by surface nonlinearity (Kauranen and Zayats 2012; Butet et al. 2015; de Ceglia et al. 2015; Celebrano et al. 2015; Finazzi et al. 2007; Dadap 2008), in high-permittivity dielectric nanoantennas the bulk nonlinearity dominates and the properties of the radiation diagram strongly depend on the nonlinear susceptibility elements, the crystallographic orientation, and the input polarization state (Finazzi et al. 2007; Dadap 2008). In this framework, the enhancement of the nonlinear response due to coupling between magnetic and electric dipole resonances has already been observed in third harmonic generation and two-photon absorption (TPA) experiments in Si nanodisks (Shcherbakov et al. 2014, Shcherbakov et al. 2015a; Shcherbakov et al. 2015b). More recently, second harmonic generation (SHG) with an efficiency of 10^{-3} has been theoretically predicted by exploiting the magnetic dipole resonance in AlGaAs structures (Carletti et al. 2015). So far, research in this field has focused on the enhancement of the nonlinear response of nanostructures rather than on the control of the radiation pattern of the nonlinearly generated signals. For example, in (Carletti at al. 2015) a very high SHG efficiency is reported, but the SH signal emitted by the AlGaAs nanodisk has a null along the cylinder axis in the forward and backward directions. Engineering the radiation pattern of SHG for e.g. achieving uni-directional signal emission is of paramount importance for using all-dielectric nanoantennas in applications

Proceedings 30th European Conference on Modelling and Simulation ©ECMS Thorsten Claus, Frank Herrmann, Michael Manitz, Oliver Rose (Editors)
ISBN: 978-0-9932440-2-5 / ISBN: 978-0-9932440-3-2 (CD)

such as chemical or biological sensing requiring low power and low cost components, (Albella et al. 2014) because it affects the collection efficiency of experiments in real-life conditions. Although the radiation pattern of SHG in centrosymmetric nanoparticles (Bennemann 1998; Dadap et al. 1999; Mäkitalo et al. 2014; Bautista et al. 2012) and THG in amorphous dielectric particles has been studied, shaping of the SHG emission from non-centrosymmetric nanoparticles exhibiting both magnetic and electric multipole resonances remains almost unexplored. The main goal of this paper is to describe a route based on the structuring of the substrate (Lezec et al. 2002; Garcia-Vidal et al. 2003; Yu et al. 2008; Iwaszczuk et al. 2013) to engineer the radiation pattern of the second harmonic (SH) signal generated by AlGaAs on aluminum oxide all-dielectric nanoantennas. The SH beam divergence is minimized by coherent forward and backward scattering of the radiation emitted at grazing angles from the optical antenna toward a concentric grating structure, whereas the symmetry of the mode is converted by introducing a suitably-designed phase shift. The parameters of the structure are optimized through extensive numerical simulations and design guidelines for fabrication are provided.

SECOND HARMONIC GENERATION IN AlGaAs NANOANTENNAS

In order to demonstrate the control of the radiation profile of the SH field generated by AlGaAs nanoantennas we use frequency-domain simulations implemented using the finite-element-method in COMSOL. The pump beam at the fundamental frequency is assumed to be a plane wave s-polarized along one of the crystalline axes, which are assumed to be aligned with the simulation Cartesian coordinate system axes (see Fig. 1). The second-order nonlinear susceptibility tensor of AlGaAs has only elements of the type $\chi^{(2)}_{ijk}$ with $i \neq j \neq k$ (Carletti et al. 2015). Thus the *i-th* component of the nonlinear polarization at the SH frequency 2ω is given by:

$$P_i^{(2\omega)} = \varepsilon_0 \chi_{ijk}^{(2)} E_j^{(\omega)} E_k^{(\omega)} \qquad (1)$$

where ε_0 is vacuum dielectric constant and $E_j^{(\omega)}$ is the *j-th* component of the electric field at the pump frequency ω. The nonlinear polarization in Equation (1) is used to define the nonlinear source currents and calculate the SHG from the AlGaAs cylinders. The reference structure that we have considered here is borrowed from (Carletti et al. 2015) and is an $Al_{0.18}Ga_{0.82}As$ cylinder with radius $r = 225$ nm and height $h = 400$ nm on top of an Al_2O_3 substrate.

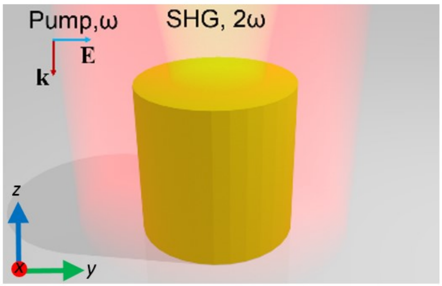

Figure 1: Schematic representation of SHG from a cylinder. The pump beam at ω is a plane wave with wave vector, k, parallel to the cylinder axis.

In order to model the dispersion of the refractive index of $Al_{0.18}Ga_{0.82}As$ we used the analytical model proposed by Gehrsitz (Gehrsitz et al. 2000) which was derived from comparison with measurements. The scattering efficiency (defined as $Q_{sca} = C_{sca}/\pi r^2$ where C_{sca} is the scattering cross-section and r is the cylinder radius) at wavelengths close to the magnetic dipole resonance is reported in Fig. 2(a). It is possible to notice that due to the presence of the substrate the magnetic resonance redshifts ($\lambda = 1655$ nm) with respect to the case of cylinder suspended in air ($\lambda = 1640$ nm).

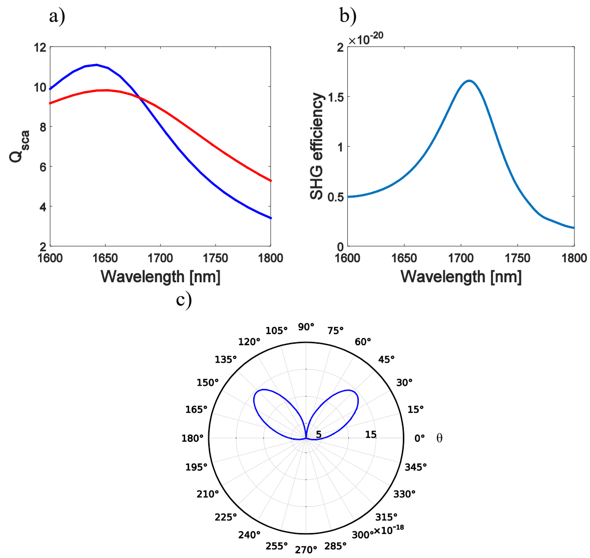

Figure 2: (a) Scattering efficiency Q_{sca}, as a function of wavelength calculated for $r = 225$ nm and $h = 400$ nm, in the case of presence of the substrate (red line) or completely air-surrounded nanostructure (blue line). (b) SHG efficiency as a function of pump wavelength for the same cylinder in the presence of the Al_2O_3 substrate and pump intensity I_0=1.33 10^{-3} W/m^2. (c) Far-field radiation pattern of the SH electric field for pumping wavelength $\lambda = 1710$ nm, y-z plane. The incident light is a plane wave with a wave vector, k, parallel to the cylinder axis and the electric field, E, polarized along the y-axis.

We investigate the SHG phenomenon by using the nonlinear polarization induced by the nonlinear susceptibility $\chi^{(2)}$ as a source in COMSOL simulations. We define the SHG efficiency as:

$$\eta_{SHG} = \frac{\int_A \vec{S}_{SH} \cdot \hat{n} da}{I_0 \times \pi r^2} \quad (2)$$

where $\vec{S}_{SH}$ is the Poynting vector of the SH field, $\hat{n}$ is the unit vector normal to the surface A enclosing the antenna and I_0 is the incident field intensity (I_0 = 1.33 10^{-3} W/m^2 in the simulations). We observed that the SHG efficiency peak is for a pumping wavelength of λ = *1710* nm, as reported in Fig. 2(b). Fig. 2(c) shows the far field in the air region at that pumping wavelength. The mode has its twofold symmetry directly transferred to its emission pattern with maximum emission intensity at large off-axis angles; the radiation null in the forward direction (θ = *90°*) directly comes from the symmetry of the nanoantenna and the relative orientation between the crystallographic axes and the input polarization state of the pump (Iwaszczuk et al. 2013).

In order to avoid the presence of this null of the radiation diagram one can take advantage of structured pump light beams or resort to different particle geometries; the approach we have considered here explores the use of structuring the substrate similarly to what is done at radio frequency where collimation is achieved through the patterning of the ground plane (Lezec et al. 2002; Garcia-Vidal et al. 2003).

Our beam collimator (described in the next Section) was derived from the plasmonic collimator that was introduced by Yu and Iwaszczuk (Yu et al. 2008; Iwaszczuk et al. 2013) and is essentially based on an interference effect in the near field: as schematically described in Fig. 3, the SHG source (the AlGaAs nonlinear cylinder) couples to the half-ring pattern acting as a 2D ensemble of scatterers that coherently radiate the energy of the SH into the far field. The design of the beam collimator, which is treated in the next section, is thus of utmost importance to shape the multipolar emission pattern of the AlGaAs nanoantenna into a uni-directional beam.

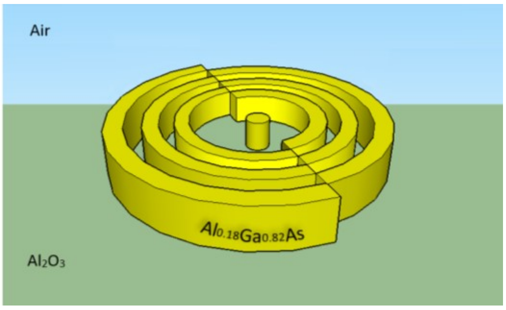

Figure 3: Schematic representation of the AlGaAs concentric grating over the Al_2O_3 substrate.

DESIGN OF THE BEAM COLLIMATOR

In this section we tackle the problem of engineering the radiation pattern of the second harmonic (SH) signal generated by the AlGaAs nanoantennas described in the previous paragraph. We demonstrate that the SH beam divergence can be minimized and the main radiation lobe can be tilted along the cylinder axis by using a concentric grating grown on the aluminum oxide substrate. Our approach mimics the surface plasmon polariton (SPP) assisted scattering from a periodic structure with the groove spacing, width and depth that are optimized for our specific geometry. Indeeed, Garcia-Vidal et al. have previously shown that it is possible to collimate and even focus the output of a subwavelength emitter, with an optimized performance for a finite number of grooves in the grating structure (Garcia-Vidal et al. 2003). This principle was later used by Yu (Yu et al. 2008), who fabricated linear and concentric arrays of grooves in the end facet of QCLs for collimation of the output. Agrawal and Nahata used concentric corrugations for resonant enhancement of transmission of THz waves through subwavelength apertures (Agrawal et al. 2005; Agrawal and Nahata 2006) and enhanced coupling between free-space THz beams and wires (Agrawal and Nahata 2007). Fig. 3 illustrates the basic design considered here. In our case we place *N* concentric corrugations with radial distance *d*, width *a*, and depth t_g around the optical antenna. The first step is to design an effective grating for the SH wavelength. We start from a 2D model of the grating. Fig. 4(a) shows the scheme of such a structure that consists of teeth composed of of high (AlGaAs) and low (air) index material deposited onto a low-index layer (Al_2O_3). The design parameters for the structure include the grating period (Λ), the grating thickness (t_g), and the duty cycle (DC). The DC is defined as the ratio of the width of the high index material with respect to Λ (Finazzi et al. 2007). Here we assume an infinite thickness of the low-index layer under the grating (t_L). We fix t_g equal to the height of the cylinder. By using the reciprocity principle, we excite the structure with a Gaussian beam at a wavelength of 855 nm (i.e. the wavelength at which the SHG efficiency should be maximum) at normal incidence and with the electric field polarized along the y-axis, and we measure the power that is scattered from one side of the grating towards a numerical probe (depicted by the red line in Fig. 4(a)). We vary the DC and the period of the grating in order to find the values that maximize the lateral scattered power. In our simulations the probe is about 1200 nm far from the grating. We repeated the simulation for different distances of the probe but we found that the result is independent from the distance.

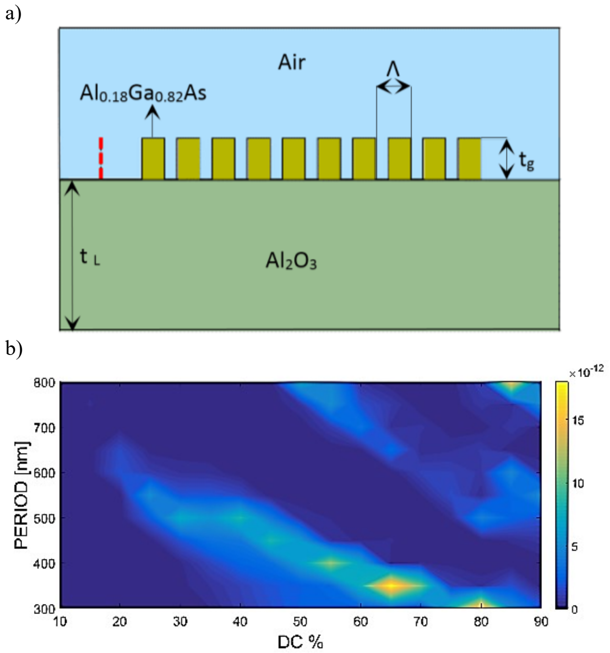

Figure 4: (a) Schematic picture of the system under study: one-dimensional high contrast grating realized in AlGaAs over an Al_2O_3 low-index layer. (b) Scattered power as a function of grating period Λ and duty-cycle for a probe placed 1200 nm far away from the grating.

We discovered that the values that maximize power are a grating period Λ = 350 nm with a DC equal to 65% that corresponds to a width of the high index material of 227.5 nm. The next step of our design procedure consists in evaluating the optical response of the nanoantenna surrounded by the grating by using a three-dimensional model. In particular, we placed *N* concentric grooves around the cylinder at a radial distance *d = 380* nm. That distance is chosen such that it does not affect the linear scattering behaviour of the cylinder at the fundamental frequency. The concentric grating structure will lead to a vertical redirection of the radiation emitted in the xy-plane; however, the quadrupole symmetry of the SHG mode still prohibits that radiation is coupled onto the normal propagation direction. Therefore, as schematically illustrated in Fig. 5, we must introduce a phase shift between the light scattered from each of the two half-rings of the concentric grating structure (see Fig. 5(a)). This will lead to a conversion of the quadrupole mode to a dipolar mode, and thus the far-field pattern will be coupled onto the propagation direction normal to the surface (Dadap 2008). Looking at the magnitude of the far-field in the forward propagation direction ($\theta = 90°$), we notice that the optimal shift between the two half rings is *s = 250* nm, see Fig. 5(c).

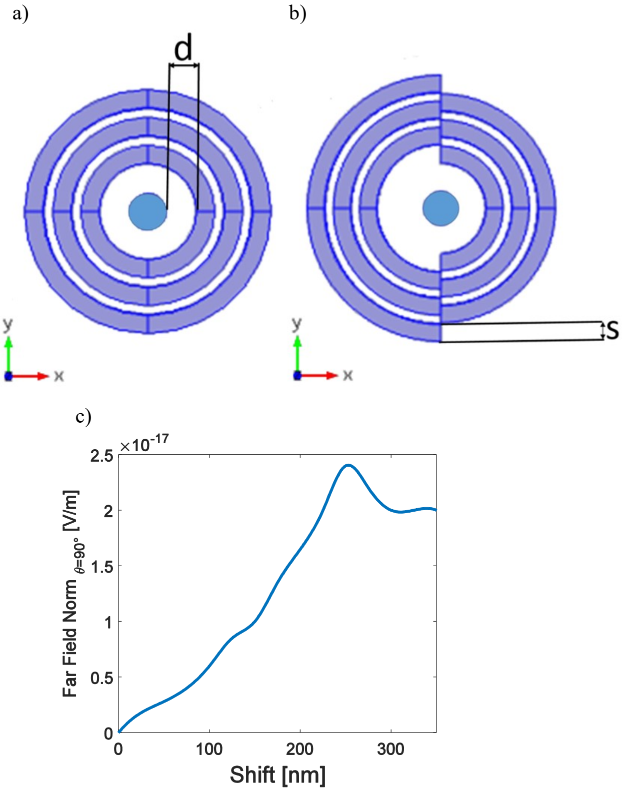

Figure 5: (a) Schematic illustration of a concentric grating structure with radial distance *d = 500* nm; top view. (b) Concentric grating with phase-shift *s = 250* nm. (c) Magnitude of the far-field in the forward direction ($\theta = 90°$) as a function of the phase shift *s*.

The effect of the grating can be verifeld by looking at the electric field above the cylinder. As it can be seen from Fig. 6(b), when the optimized grating is present the zero at the normal direction ($\theta = 90°$) disappears. Fig. 6(c) shows the calculated far field intensity pattern emitted from the structure with the *N = 3* grating. We observe a clear collimation of the generated SH field into a narrow, forward-propagating lobe. The asymmetry observed in the emission in the *x-z* plane from the optimized structure is due to the asymmetry introduced in the grating structure in that plane, combined with the usage of a grating with finite number of grooves. We have observed (results not shown here) that, by increasing the number of grooves, this asymmetry is significantly reduced. However, we point out that our interest is in the realization of ultracompact frequency converters operating at the nanoscale, thus we aim at finding a good trade-off between size and performance. Moreover, the simulation of structures with high N is an extremely demanding task and therefore, for the present analysis, we limit ourselves to small values of N.

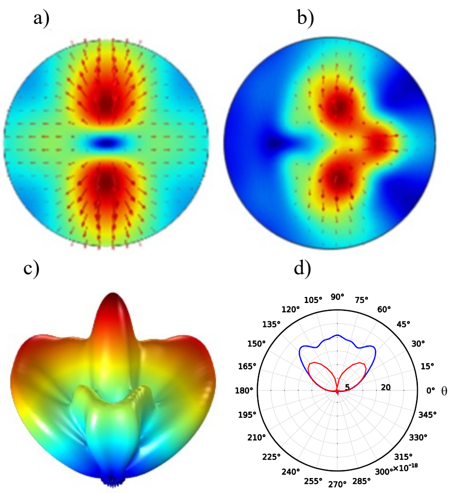

Figure 6: Magnitude of the SH electric field in the *x-y* plane (top view) in the case of, (a), single cylinder, and, (b), $N = 3$ grating. (c) SH far field in the *y-z* plane in the case of $N = 3$ grating. (d) Comparison of SH radiation pattern in the air region for pumping wavelength $\lambda = 1710$ nm in the case of single cylinder (red line) and $N = 3$ grating (blue line), *y-z* plane.

The Second Harmonic Generation efficiency as a function of the pump wavelength for the structure composed of the nanoantenna surrounded by the grating is shown in Fig. 7(a). We can observe a maximum at $\lambda = 1655$ nm, which corresponds to the magnetic dipole resonance of the cylinder alone (see Fig. 2(a)). This may be due to the fact that the structure formed by the cylinder with concentric grating around it has a strong resonance at that wavelength. In addition, the efficiency when the grating is present is slightly higher with respect to the case of the isolated cylinder structure.

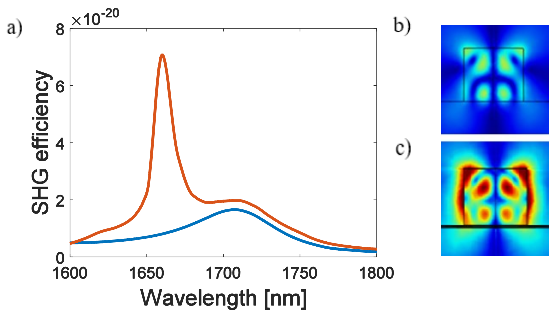

Figure 7: (a) Comparison of SHG efficiency in the case of single cylinder (blue line) and $N = 3$ grating (orange line). (b) Normalized $|E^{SH}|$ on a cross section in the *y-z* plane at the centre of the cylinder without grating at pumping wavelength $\lambda = 1710$ nm. (c) Normalized $|E^{SH}|$ on a cross section in the *y-z* plane at the centre of the cylinder with $N = 3$ grating at pumping wavelength $\lambda = 1710$ nm.

CONCLUSION

We report the design of an AlGaAs nanoantenna based on a nanodisk with a concentric grating structure that guarantees a desired SH radiation pattern. We show that, by careful engineering of the surrounding grooves, it is possible to convert a radiation pattern with azimuthal mode number $m = 2$ into a dipolar-like emission profile. This was accomplished by introducing a concentric grating structure that scatters the SH radiation in the forward direction. The symmetry of the generated SH mode was broken by an engineered asymmetry of the grating structure, leading to a highly collimated output.

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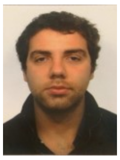

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