Strong enhancement of second-harmonic generation in all-dielectric resonant waveguide grating

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The authors demonstrate that a dielectric resonant waveguide grating can enhance optical second-harmonic generation by a factor of 550 compared to a similar flat surface. Their structure, which consists of purely dielectric and thereby transparent materials, has a low index silicon dioxide grating covered by a high index titanium dioxide layer and it is optimized for the fundamental wavelength of 1064 nm. Polarization dependence of the second-harmonic response suggests that the enhancement arises from the favorable interaction of the resonant waveguide mode and its strong local field with the inherent surface nonlinearity of the dielectric materials. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2783969]

The electromagnetic field of light can be significantly modified locally by properly designed nanostructures.[1] Often the goal of locally enhancing the field is either to reduce the area under study below the wavelength scale (e.g., near-field microscopy[2]) or to strengthen the process of interest (e.g., surface-enhanced Raman scattering[3]). The local field enhancement is also very important for nonlinear optical effects, which scale with a high power of the field.[4] Redistribution of radiative energy can give rise to such large local enhancement of the nonlinear response that even the response averaged over the whole sample is significantly enhanced.

A common way to localize and intensify electromagnetic fields is to excite plasmons on metal surfaces or particles. Surface plasmons enhance second-harmonic generation (SHG) from solid films by several orders of magnitude.[5] Similarly, metal nanoparticles arranged in ordered patterns can give rise to a nonlinear response due to particle plasmons, which can be improved by introducing additional features such as gaps between the particles.[6] Less ordered features such as varying the roughness of metal surfaces,[7] random semicontinuity[8] of the films, and grains of metal on insulating surface[9] can also lead to high enhancement. However, metallic losses can be problematic when the sample is illuminated by a bright laser beam or transmitted light needs to be detected.

A resonant waveguide grating[10] (RWG) consists of a dielectric grating whose period is shorter than the wavelength of light and is covered by a thin coating of a high refractive index material. A RWG simultaneously acts as a waveguide and couples externally propagating light at a suitable angle of incidence into a waveguide mode, which is associated with a strongly enhanced local field. The same grating also couples light out of the waveguide. As a consequence, a narrow angular and spectral region of high reflectivity occurs. More detailed properties of the RWG structures are described elsewhere.[10,11] In practice, RWG needs to be optimized using numerical computations. In linear optics, they can be useful to study, e.g., biomolecular interactions.[12] In nonlinear optics, a double-layer guided-mode resonance filter coated with an active nonlinear material increased second-harmonic (SH) signal by a factor of 9.[13] More lately, over 300-fold enhancement of two-photon fluorescence (TPF) was demonstrated in a resonant double grating waveguide structure functionalized with an organic molecule.[14] Both of these structures are similar to the RWG.

Other nonlinear experiments taking advantage of gratings without metals have also been performed. Blau *et al.* achieved counterpropagating phase matching for SHG in organic waveguides with gratings.[15] Pezzetta *et al.* achieved phase matching by combining grating and periodic modulation of the nonlinearity of the material in a waveguide.[16] Lagugne-Labarthet *et al.* enhanced SHG using an electrically poled surface relief on a thin film of an azopolymer.[17] Although dielectric structures avoid metallic losses, the enhancement of the nonlinear response has generally been rather modest compared to plasmon-assisted solutions with the exception of the TPF experiments such as those in Ref. 14. This is because the structures have earlier been optimized to achieve phase matching or to use certain active material, whereas we aimed to optimize the enhancement by maximizing the field amplitudes.

In this letter, we report an enhancement of SHG by a factor of over 550 from a fully dielectric RWG without active molecules or metals. The enhancement is found to be limited to a very narrow region of angle of incidence where the resonant mode is excited. The polarization dependence of the response suggests that the enhancement is due to the favorable interaction of the resonant mode with the inherent surface nonlinearity of the dielectric material.

The design of our RWG sample (Fig. 1) was based on rigorous diffraction theory[18] and numerical calculations. The structure has TiO$_2$ as a high index of refraction material on top of a SiO$_2$ grating with lower index of refraction. The grating was optimized for the fundamental wavelength of 1064 nm (laser used in the experiments) and rather low angle of incidence of about 20°. The local field enhancement was optimized for *p*-polarized fundamental radiation, which is expected to couple favorably with the sample's surface nonlinearity that has a strong out-of-plane character. An interest-
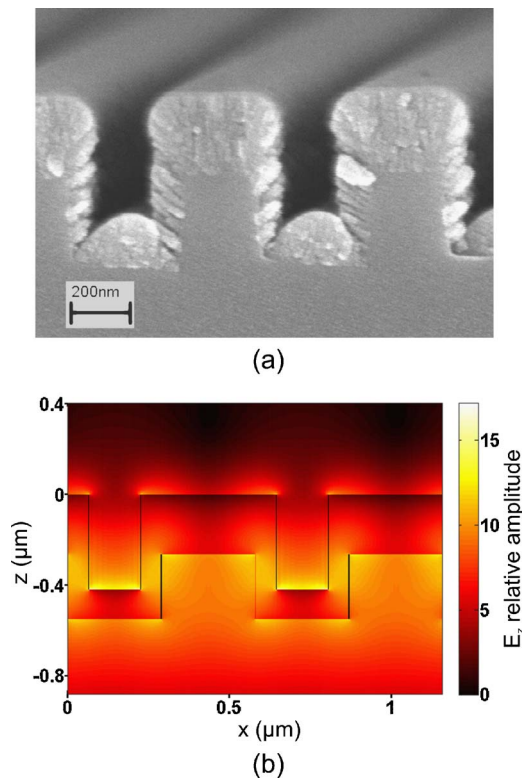
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FIG. 1. (Color online) (a) SEM image of the RWG sample. (b) The calculated amplitude of the electric field component along the sample surface normal (E_z) in the vicinity of the periodic RWG structure at resonant conditions.

ing detail is that the calculations predict strongest local fields at slightly larger angle of incidence than the maximum reflectivity.

The SiO$_2$ grating with period of 580 nm, depth of 287 nm, and fill factor of 0.5 was fabricated using electron-beam lithography and reactive ion etching. 262 nm of TiO$_2$ was added with vacuum deposition. The actual thicknesses of the TiO$_2$ coating on the bottom of the grooves and on the sidewalls are 50% and 25% of that on top of the grating, respectively, due to the used evaporation process. The effects of this nonuniformity were taken into account during the design step. A scanning electron microscope (SEM) image of the sample is shown in Fig. 1(a) and the calculated electric field in Fig. 1(b). The numerical calculations predict that the amplitude of the electric field can be enhanced by a factor of up to 17 at the bottom of the grooves compared to the incident plane wave.

Proper operation of the RWG was verified by linear optical measurements. The sample was illuminated using a *p*-polarized laser beam at the wavelength of 1064 nm. Both reflectance and transmittance of the RWG were recorded as functions of the incidence angle. The exact location of the experimentally observed resonance differs from theoretical simulations, but by tuning the simulation parameters with values obtained from the SEM image in Fig. 1(a) (depth of 290 nm, coating of 232 nm with 33% thickness on the bottom of the groove and 27% on the sidewalls) the theoretical resonance moves from 20.8° to 22.2°, as shown in Fig. 2. Experimentally, measurements seen in Fig. 2 give about 0.25° wide resonance peaks at 22.4°, very close to the theoretical value. The measured width of the resonance is comparable to the divergence of the laser beam, which therefore
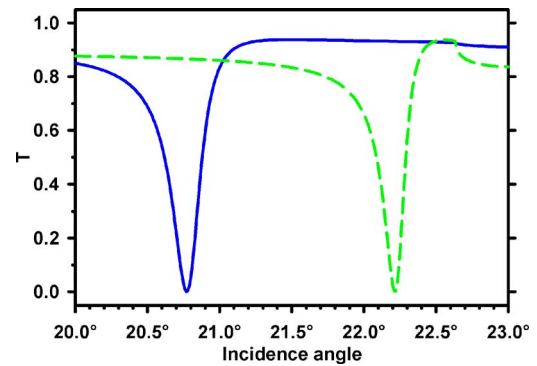

FIG. 2. (Color online) Theoretical transmittance (T) of the optimized RWG as a function of the incidence angle using the original design parameter values (solid blue line) and values corrected using the SEM image in Fig. 1(a) (green dashed line). The back surface of the glass substrate is ignored. Therefore the reflectance (R) is $R = 1 - T$ and is not shown.

affected the width and magnitude of the measured reflectance peak.

In the nonlinear measurements, the sample was illuminated with a pulsed Nd doped yttrium aluminum garnet laser (wavelength of 1064 nm, pulse length of 70 ps, and pulse energy of 0.1 mJ). Great care was taken to collimate the beam as well as possible to enable efficient coupling into the resonant mode. The fundamental beam incident on the sample was linearly polarized using a Glan polarizer and filtered with a glass filter to remove any SH light generated before the sample. The SHG signal was detected in the transmitted direction with a photomultiplier tube after filtering out the fundamental beam and choosing the detected polarization by another Glan prism. We used a narrowband interference filter and checked the quadratic dependence of the signal intensity on fundamental beam power to further verify that only SHG was being detected.

The *p*-polarized SH light from the RWG as a function of the incidence angle of a *p*-polarized fundamental beam is shown in Fig. 3. The resonant enhancement of the SHG is clearly visible as a narrow peak centered at about 22.5° of incidence. This is 0.15° higher than the location of the maximum reflectance, in qualitative agreement with the theoretical calculations. By varying the polarization states of the fundamental and SHG beams, it was verified that the enhanced SHG signal corresponds to linear *p* polarization for both incident and detected light. If either was *s* polarized, no
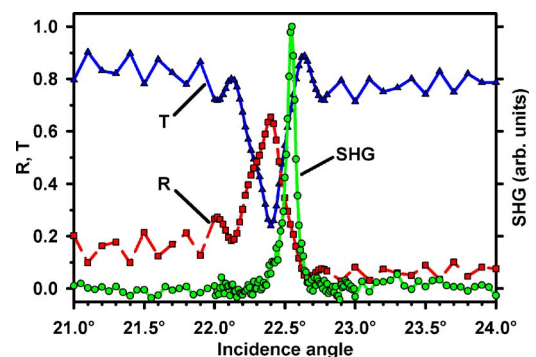

FIG. 3. (Color online) Measured dependence of the transmittance T (triangles with blue solid line), reflectance R (squares with red dashed line), and *p*-polarized SH light (SHG) (circles with green solid line, narrow peak) on the angle of incidence of the fundamental beam. Note that SHG is given in arbitrary units and peaks at a slightly higher angle of incidence than R.

signal could be observed. Note that only *p*-polarized incident light, which is enhanced, can give rise to local field components along the local surface normal, whereas local field components due to incident *s*-polarized light always lie in the local surface plane of the structure. Therefore, only *p*-polarized incident light can interact with the local surface nonlinearity of the structure. Furthermore, the symmetry of the local surface nonlinearity is such that it can only give rise to *p*-polarized SH light for *p*-polarized incident light.[4] The physical explanation for the SHG enhancement is therefore the enhanced amplitude of the fundamental electromagnetic fields at the dielectric interfaces due to excitation of the resonant mode and their interaction with the inherent surface nonlinearity of the dielectric material.

Because of the limited sensitivity and dynamic range of photomultiplier tubes, additional steps were needed to calibrate the enhancement. A planar TiO_2 coating had only very small effect on the magnitude of SHG from clean SiO_2 and thus we used the coated SiO_2 surface as our reference. In order to have a measurable SHG from a planar SiO_2 reference without the grating, we had to focus the fundamental beam using a 10 cm focal-length lens. Unfortunately, the greatly increased divergence of the laser beam due to focusing leads to wider distribution of wavevectors in the fundamental beam which do not all match the narrow resonance. This results in a significant reduction of the SHG response of the RWG. Therefore, we used two samples of organic nonlinear molecules (terthiophene-vinylbenzoate) known to form homogeneous and isotropic Langmuir-Blodgett films as intermediate references (labeled A and B) between the collimated and focused measurements.[19]

Using the focused setup, the SHG from the TiO_2 reference was compared to the SHG of the organic samples having a calibrated neutral density filter in the fundamental beam before them. The measured SHG intensity ratio using intermediate references A and B was 1:3.27 and 1:2.68, respectively. The filter transmitted 4.1% of the fundamental beam, which corresponds to the ratio of 1:595 in the SHG signals with and without the filter. Then the collimated setup was used to compare the level of SHG from the intermediate references and the RWG at resonance. This yielded signal ratios of 1:0.290 and 1:0.385 using references A and B, respectively. As a result, the SHG intensity measured from the RWG at resonance compared to the smooth TiO_2-coated glass surface is higher by a factor in the range of 564–614. The variations are believed to be due to small imperfections of the organic references. Therefore, we conclude that the SHG enhancement of our RWG is at least 550 or better.

The intensity of the SH signal scales with the fourth power of the amplitude of the fundamental field, ideally allowing an enhancement of at least four orders of magnitude in our case. However, only certain regions of the sample where the enhanced fundamental fields and nonlinear interactions occur contribute to the enhancement. Nevertheless, the present results, albeit very promising, are still far from the theoretical limit for enhancement. We therefore believe that further improvements in the sample design and fabrication could lead to even higher enhancements in the nonlinear response.

In summary, we have shown that a purely dielectric, unfunctionalized, and nonabsorbing RWG structure consisting of SiO_2 and TiO_2 enhances the SHG signal by more than a factor of 550 in comparison to a similar surface without the grating. The dependence on the angle of incidence and polarization properties of the SHG shows the enhancement results from the resonant mode interacting with the inherent surface nonlinearity of the dielectric material. Therefore, the enhancement must be due to the strong local fields at interfaces, not changes in the nonlinearities of the materials. We believe that by optimizing and perfecting the structure even higher enhancements are possible paving the way for interesting applications utilizing the sensitivity of the structure in, e.g., biomolecule detection.

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