Giant enhancement of second-harmonic generation in multiple diffraction orders from sub-wavelength resonant waveguide grating

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Abstract: We demonstrate a purely dielectric resonant waveguide structure that enhances the efficiency of second-harmonic generation by a factor of at least 5500 compared to a flat reference surface in the same geometry. We also show that the structure emits second-harmonic radiation in four different directions when the sample is illuminated with fundamental radiation incident at the resonant angle of the sample.

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1. Introduction

Structural details that are smaller than wavelength of light can give rise to strongly enhanced local electric fields and very sharp resonances. Resonant waveguide gratings (RWG) [1,2] constructed from all-dielectric, low-loss materials are one example of such sub-wavelength structures. A RWG consists of a surface waveguide, which is lossy due to a surface grating diffracting light both in and out of the waveguide mode. On resonance, the fields diffracted out of the waveguide mode interfere with the transmitted and reflected fields in such a way that it is theoretically possible to achieve 100% reflection [1]. Simultaneously, the field strengths in the waveguide mode can be significantly enhanced. RWGs can thus provide efficient and convenient coupling of freely propagating laser beams to nanosized structures and waveguide modes while avoiding problems such as those arising from tight focusing.

Due to their unique characteristics, RWGs are commonly used as narrow bandpass filters or selective mirrors [3–7]. In filter use, the strongest local fields are often inside the grating material and the enhanced field is not used directly. Instead, an effort to minimize the enhancement can be beneficial to, e.g., decrease the damage threshold [8]. However, designing the RWG using, e.g., deep grooves so that the local field is both strong and accessible at the surface, can lead to applications such as improved fluorescence detection [9,10]. In particular, the high enhancement of the local fields at the surface of the RWG is interesting for nonlinear optical effects, which scale with a high field intensity, as can also occur in a total-internal-reflection geometry [11].

In this paper, we demonstrate over 5500-fold enhancement of second-harmonic generation (SHG) from an all-dielectric sub-wavelength RWG. The result is a significant improvement over the previous enhancement factor of 550 reported in Ref. 12. This is a direct consequence of careful sample fabrication and higher sample quality, which allows more efficient coupling into the structure and thereby leads to stronger local fields. The enhancement factor is evaluated using an air-SiO_2 flat interface as a reference sample. We evaluate the enhancement for a similar experimental geometry for both samples and geometries optimized for each sample. Our experimental results are found to be in agreement with the simple estimate obtained from the local-field calculation in the structure. In addition, we report that second-harmonic (SH) beams of similar intensity can be detected in four different directions, which correspond to the allowed diffraction orders of the SH light.

2. Experiments

The resonant structure is composed of a low index of refraction SiO_2 grating coated with a layer of TiO_2 as a high index of refraction material. The SiO_2 grating was fabricated using electron-beam lithography and reactive ion etching. The basic structure is similar to that used in our previous experiments [12] but the quality of the present sample was significantly improved. Specifically, the electron-beam system (Vistec EBPG 5000 + ESHR) is improved, enhancing the patterning quality of the SiO_2 grating and the evaporation process has been upgraded. Pure oxygen can now be pumped into the chamber during evaporation, allowing better control over the refractive index of the TiO_2 deposited onto the sample. Minor changes were also made in the geometrical properties of the structure, mostly because the TiO_2 coating thicknesses on the sidewalls and on the bottom of the grooves were slightly different with the new evaporator. Calculations show, however, that these changes had only a small effect in the energy densities of the local fields in the structure.

The present sample was designed for optimum coupling into the waveguide mode of *p*-polarized light at the fundamental wavelength of 1064 nm and for a resonance angle of incidence of about 21°. These parameters were chosen because the surface nonlinearity of the

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sample has a strong out-of-plane nature and the nonlinear response is therefore expected to be particularly enhanced for *p*-polarized fundamental radiation. The local field enhancement resulting from the grating resonance was thus optimized for this particular polarization on the basis of numerical calculations and rigorous diffraction theory. The theoretically calculated linear transmission as a function of the incident angle is shown in Fig. 1.

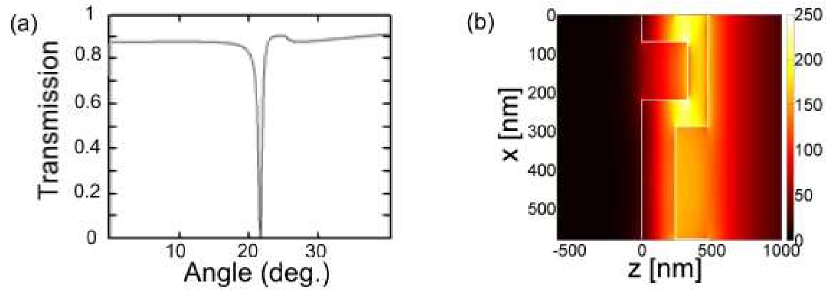

Fig. 1. (a) Theoretically calculated transmission of the RWG at 1064 nm as a function of incident angle, (b) scanning electron microscope image of the RWG, and (c) calculated time-average energy density in the RWG (delimited by the white lines) at 1064 nm.

The period of the grating is 580 nm and the depth of the grooves is 237 nm. The thickness of the TiO_2 coating is 237 nm on top of the grooves and 60% and 30% of that on the bottom of the grooves and on the sidewalls, respectively. The calculation of the local field [see Fig. 1(b)] reveals that its energy density can be enhanced by a factor exceeding 200 in certain areas of the grating structure compared to the field in free space. Although the strongest field is located at the sharp grating corners, the actual structure exhibits rounded corners, which decrease the field strength at these locations. Beyond the corners, the strongest local fields appear in the TiO_2 layer and at the air-TiO_2 interface throughout the structure. While the TiO_2 layer may give rise to quadrupolar bulk nonlinearities [13], the air-TiO_2 interface is favorable for the coupling with the surface nonlinearity arising from the broken centrosymmetry of the interface. Although one cannot separate the bulk and surface contribution to the nonlinearity, we expect the bulk contribution to be significantly smaller than that of the interfaces. The enhancement factor of 200 means that the intensity of the SH signal generated from the grating could ideally be over four orders of magnitude larger than from a flat surface. This estimation is simply based on the local field enhancement. For a more detailed analysis, one should account for the coupling of the radiation fields (at the fundamental and harmonic frequency) to the local fields, the locally varying nonlinearity and the local nonlinear sources.

Our structure has a sub-wavelength period for a fundamental wavelength of 1064 nm. Therefore, only zero diffraction orders in transmission and reflection can propagate in air, although the waveguide mode propagates within the structure. The period is, however, larger than the SH wavelength of 532 nm. As a consequence, SH light can be emitted also into non-zero diffraction orders. From momentum conservation along the surface and taking into account the additional momentum induced by the grating, we find that the possible magnitudes of the in-plane wave vectors of SH light are given by

$$n_2 \sin\theta_m = n_1 \sin\theta_i + m(\lambda / a), \tag{1}$$

where m is the diffraction order, subscripts i and m refer to the angles of incident and diffracted plane waves, respectively. The wavelength λ corresponds to that of the SH and a is the period of the grating. Here, n_1 and n_2 are the indices of refraction of the medium in which the fundamental and SH beams propagate, respectively. In the present case light propagates in air and n_1 and n_2 are equal to 1. From this it follows that only the reflected and transmitted diffraction orders $m = 0$ and $m = -1$ can propagate and we should therefore observe SH light in four different directions as illustrated in Fig. 2(a).

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A schematic of the experimental setup is shown in Fig. 2(b). The RWG was placed in a computer-controlled rotation mount which allows the SH signal to be measured as a function of the angle of incidence, thereby allowing the angle to be optimized for maximum SH radiation. The sample was illuminated with a pulsed Nd:YAG laser operating at 1064 nm and producing pulses of 70 ps duration. This fundamental beam was linearly polarized using a Glan-polarizer. Special care was taken in order to minimize the beam divergence which may decrease the coupling efficiency into the waveguide mode. The laser spot on the sample surface had a diameter of ~1 mm. The beam was subsequently filtered to remove all possible SH light that might be generated by the optics before the sample. In this way, the radiation incident onto the sample was purely at the fundamental wavelength and *p*-polarized. A photomultiplier tube for the detection of SH light was placed at the end of a computer-controlled rotating arm, thus making it possible to detect SH light over the whole 360° range. The fundamental wavelength was filtered out after the RWG and another Glan-polarizer was used to polarize the SH light along the *p*-direction before detection with a photomultiplier tube. The detector area was limited by a 200 µm wide slit, which is significantly narrower than the beam diameter at the detector allowing for better resolution of the SHG signals.

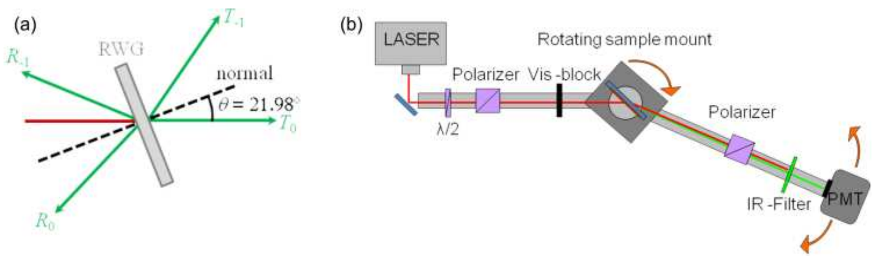

Fig. 2. (a) Directions of the SH light diffracted by the RWG. T_0 (T_{-1}) and R_0 (R_{-1}) represent the directions of the transmitted and reflected 0 (−1) order. The direction of the incident fundamental beam is marked in red and the normal to the surface with the dashed line. (b) Experimental setup. $\lambda/2$: half-wave plate, Vis-block: filter blocking any visible radiation from the laser, IR-filter: filter blocking any IR radiation from the RWG, PMT: photomultiplier.

3. Results and discussion

We first determined the resonance angle for maximum SH enhancement by detecting the transmitted 0 diffraction order while changing the angle of incidence. The resonance was found to occur at the 21.98° angle of incidence, close to the design target of 21°.

In order to estimate the enhancement factor in SHG induced by the RWG we used a planar SiO_2 surface as a reference. This is different from the air-TiO_2-SiO_2 structure of the grating. However, we have verified experimentally that the nonlinear response from the air-SiO_2 interface and the air-TiO_2-SiO_2 structure had a negligible difference, although both the air-TiO_2 and TiO_2-SiO_2 interfaces can coherently contribute to the nonlinear response of the latter structure. For the experimental conditions described in the above section, the SHG signal produced by the planar SiO_2 surface, however, was extremely small. We therefore had to focus the fundamental beam on the planar surface in order to reliably measure the SH signal. The beam was weakly focused with a 10 cm lens resulting in an effective 0.005 numerical aperture. The SH signal from the reference sample thus diverges weakly and was easily captured by the detector. On the other hand, for the case of the RWG, a focused beam reduces the coupling efficiency into the waveguide mode as the wide distribution of the wavevectors of the focused fundamental beam do not match the narrow resonance of the grating. In order to compare the results obtained using the collimated (RWG grating) and focused (SiO_2 surface) setups, an intermediate sample was used as a reference. The intermediate sample was an organic thin film of nonlinear terthiophene-vinylbenzoate (TSe) molecules [14]. The enhancement factor in SHG produced by the RWG compared to the flat surface was then performed in 3 steps. First, the SHG signal ratio produced by the flat SiO_2 reference surface

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and the intermediate sample was measured in the focused configuration and was found to be in the range of 1:7.55-1:10.0, where the variation was related to the spatial inhomogeneity of the TSe sample. Note that due to the large SHG signal emitted by this intermediate sample in the focused configuration, a calibrated filter was inserted in the setup. The filter transmitted 8.3% of the fundamental beam, which corresponds to the ratio of 1:145 in the SHG signals with and without the filter. Finally, the collimated setup was used to measure the SHG signal ratio between the intermediate sample and the RWG in resonance and found to be 1:5.17. As a final result we can estimate the SHG intensity produced by the RWG in resonance to be larger than that produced by the planar TiO_2 surface by a factor in the range of 5500-7500. This rather remarkable result exceeds by an order of magnitude the previous reported results [12] reflecting the substantially higher quality of the present sample.

The enhancement factor given above was estimated with respect to a planar SiO_2 surface assuming an angle of incidence identical to that of the RWG resonance. Although this is a natural choice, the rather small angle of incidence of about 22° leads to a relatively weak SH response from the planar reference due to the strong out of plane character of the surface nonlinearity. One could therefore argue that the enhancement factor is overestimated. Note that in our previous results [12] this dependence was not accounted for in the estimation of the enhancement factor. Using the values of the SH tensor components of glass that can be found from literature [13], the maximum intensity of the SH signal emitted by the flat reference surface is estimated to occur at the incident angle of 68.5° with a ~6-fold intensity increase compared to the 22° angle. Consequently, even if we consider the optimized configuration for the planar reference the enhancement factor of the RWG for SHG is still close to 1000.

We subsequently investigated the directions in which SH light is emitted by the RWG sample and compared them with those predicted by diffraction theory. For this purpose, the detection arm was rotated over the whole 360° range in the plane of incidence while keeping the angle of incidence fixed at the 21.98° resonance. The detected SHG intensity as a function of the detection angle is shown in Fig. 3 where we clearly observe SH radiation in four different directions corresponding to detection angles of −147.42°, −33.28°, 21.98° and 157.98°, respectively. The zero angle corresponds to transmission direction through the RWG, normal to the sample surface (see Fig. 2). Positive and negative angles indicate clockwise and counterclockwise rotation with respect to the zero angle. The SH radiations detected at negative angles correspond to −1 diffraction order, while SH radiations detected at positive angles correspond to 0 diffraction order. Assuming a resonant incident angle of 21.98°, the directions of the detected SH radiation for reflected 0 order and reflected/transmitted −1 order agree very well with the values of −147.12°, −32.88°, and 158.02° estimated from Eq. (1). The small discrepancy can be attributed to the uncertainty of the grating period. When limiting the detector area by the 200 µm slit, all the SHG beams exhibited comparable angular widths of ~0.2° at a 0.5 m distance from the sample, showing evidence of strong directionality.

We further observed an azimuthal angular dependence in the SH intensity, i.e., dependence on the orientation of the sample about the surface normal, but this dependence was only observed for the SH signals detected at negative angles, i.e., for the −1 diffraction order, which makes it impossible to unambiguously determine the relative magnitudes of all the four SH beams. Indeed, when the azimuthal angle was optimized to maximize the SH intensity detected in the −1 diffraction order, the SH signal in those directions was significantly larger than that detected in the 0-order directions for which the tuning of the azimuthal angle had little effect. Furthermore, depending on the measurement geometry, some weak side-maxima (not shown here) were observed on either both transmitted or both reflected peaks. In this case the weak side-maxima are caused typically by the working field borders in the electron-beam writing that cause additional periodicity on the structure. We believe that the azimuthal dependence of the SH intensity may be caused by unaccounted interference and diffraction effects in the RWG sample. The fact that the SH intensity

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seemingly depends on the azimuthal angle of incidence may induce a further increase in the enhancement factor if the azimuthal angle is optimized for a particular diffraction order.

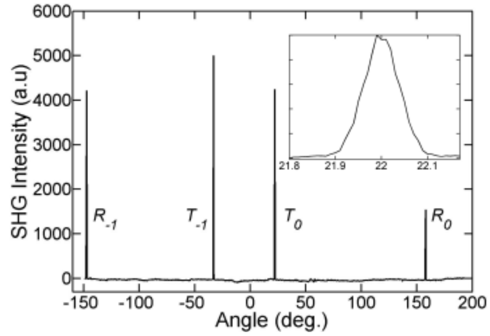

Fig. 3. Second harmonic vs. detection angle. The 0° angle corresponds to the normal to the RWG. T_0 and R_0 represent the directions of the transmitted and reflected 0 order while T_{-1} and R_{-1} represent that of the −1 order. Inset: detailed view of the 0th diffraction order in transmission.

4. Conclusions

We have demonstrated a purely dielectric resonant waveguide structure that enhances the efficiency of second-harmonic generation by a factor of at least 5500 compared to a flat reference surface in the same geometry. Even when the difference in the optimal conditions for the grating and the reference are taken into account, the enhancement factor is about 1000. The enhancement is associated with strong local fields at the interfaces of the grating when the resonant waveguide mode is excited. This remarkable enhancement in second-harmonic generation is of interest for applications that uses the sensitivity of the structure such as molecular fluorescence enhancement and biomolecule detection [9,10,15]. We have further demonstrated that the present structure emits second-harmonic radiation in four different directions when the sample is illuminated with fundamental radiation incident at the resonant angle of the sample. The additional signals arise from the fact that the period of the structure is sub-wavelength only for the fundamental wavelength used, but not for the second-harmonic wavelength. The relative magnitudes of the four peaks were found to be very sensitive to minute variations in the measurement geometry and merit further investigations.

We emphasize that the present results were obtained using a structure that supports a waveguide resonance for the fundamental wavelength only. In the future, it will be interesting to design new types of structures that are simultaneously resonant for both the fundamental and second-harmonic wavelengths so as to obtain even higher enhancement factors. Finally, we point out that even higher enhancement may be expected for third-order processes, because of their higher-order dependence on the local field amplitude and because their origin is not constrained to the surface nonlinearity by symmetry.

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