Angle-dependent SHG enhancement from nanoscale double-hole arrays in a gold film

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Abstract. We present enhanced second harmonic generation (SHG) from arrays of overlapping double holes in a gold film with varying angles of incidence. The double-hole structure permits enhanced SHG when the two holes are just overlapping to produce two sharp apexes. For different center-to-center hole spacings, the maximum occurs for different angles, which is attributed to a shift in the linear transmission wavelength peak of the array and to the requirement to break centrosymmetry for SHG.

1. Introduction

In 1998, Ebbesen and co-workers showed that an array of nanoholes in a metallic film enhanced the optical transmission compared to the prediction of the Bethe’s theory [1]. Since that work, the influence of the hole-shape on the optical properties of subwavelength holes in metals has been studied [2,3]. It has been shown that the aspect ratio of elliptical and rectangular holes changes the polarization, the transmission intensity and the cutoff wavelength, both in arrays and in isolated holes. Hole-shapes that enhance the local field intensity are of interest for nonlinear optical interactions with materials, such as second harmonic-generation (SHG).

The efficiency of SHG is proportional to the square of the local field intensity. Furthermore, SHG is not possible in centrosymmetric materials such as bulk gold, but it may be observed at the surface where the symmetry is broken. It has been shown experimentally and theoretically that the enhanced local field from rough surface and metallic nanostructures enhances the SHG signal [4]. Individual surface defects on a metallic surface [5], or nanoscopic metal tips [6] have been studied using SHG. SHG enhancement has been already measured on randomly distributed small holes [7], periodic nanohole arrays in a metallic film [8], and periodically nanostructured metal film consisting of a single subwavelength aperture surrounded by concentric circular grooves [9]. The SHG from arrays of single holes has been shown to depend on the angle of incidence. For normal incidence, where the array was centrosymmetric, negligible SHG signal was observed as expected.

Previously, we proposed a double-hole design which permits an enhancement of the local field intensity [10]. This structure consisted in two holes which were overlapping to produce two apexes. These two apexes are responsible for the extreme subwavelength focusing of this structure. We have already shown the enhancement of SHG as compared to the circular and the oblate hole structures [10]. In this paper, we present SHG angle dependence measurements from this double-hole structure.
2. Experimental techniques.

2.1. Nano-fabrication

Figure 1 shows a scanning electron microscope image of an array of double-holes milled by a focused ion beam through a 100 nm thick gold layer with a 5 nm Cr adhesion layer on a glass substrate. The gallium ion beam was set to 30 keV for milling with a current of 100 pA. The typical beam spot size was 10 nm. The dwell time of the beam at 1 pixel was 3 ms and the average time taken to mill one array of several thousands of holes was 2 minutes. Each array covers a 25×30 µm² area. We fabricated two runs of arrays. The first run varied only the spacing between the two holes, with constant periodicity. The second used a fixed spacing between the two holes but with varying the periodicity from 380 nm to 750 nm. Only results from first run will be described here.

2.2. Optical linear measurements

The linear optical transmission measurements were taken with a microscope setup using a 100× microscope objective. The transmission of white light through the array was collected using a 400 µm core broad-area fiber placed 3 cm away from the sample. The spectra were acquired using an optical spectrum analyzer. A polarizer was placed before the collection fiber to obtain the y-polarization as defined with respect to the basis and array on Figure 1. The x-polarization had at least an order of magnitude lower transmission signal.

2.3. SHG measurements

Figure 2 shows the SHG setup. SHG measurements were performed by using a Ti:Sapphire laser for delivering a 30 fs pulse. The average power incident on the sample was limited to 20 mW. A 20× microscope objective focused the pulse onto the array. The transmitted signal was collimated by a 20× microscope objective. The sample was rotated from 0° to 20° with a precision of 2°. Two BG40 filters were used to reduce the fundamental beam by 10¹⁴, at the expense of 40% of the SHG signal. The SHG was measured by a streak camera operating in synchroscan photon counting mode. As expected, SHG was only observed when the Ti :Sapphire laser was in pulsed operation, due to the requirement for high peak power. Furthermore, the pump-power dependence of the SHG signal gave a 1.99 slope on a log-log plot; almost exactly the expected value of 2 from SHG.

3. Results and discussion

Our previous work presented the enhancement on SHG from double-hole arrays as a function of the hole-to-hole spacing [10]. The enhancement of SHG was directly connected to the local enhancement of the electric field at the apexes when the holes were just overlapping. A large enhancement was measured for the y-polarization and nearly no signal was measured for the x-polarization. In that work, there was only a single non-normal angle of incidence was used to break the centrosymmetry. In this paper, we study the dependence of the SHG on the angle of incidence.

Figure 3 shows the linear normal incidence transmission spectra measured for three different center-to-center hole spacing for y-polarization collection. The array periodicity was fixed at 750 nm. There are two maxima at 600 nm and 800 nm which correspond to the (1,1) and the (1,0) resonances of the gold-air interfaces. Due to the wavelength of the Ti:Sapphire laser around 800 nm, the (1,0) resonance is of most interest here.
Figure 4 shows the measurements of the SHG through the arrays for different spacing and different angles of incidence. The general trend of this figure is that the holes that are nearly overlapping produce the greatest SHG, which is maximized for an angle of 8°.

For the angles smaller than 5°, the SHG was weak. The maximum of signal measured was 133 counts. For such small angles, the centro-symmetry of the sample is not broken sufficiently, which is a necessary condition for measuring a significant SHG signal. A previous work reported that a 10° angle was necessary angle to measure any significant signal from an array of circular and triangular holes in a gold film [8].

By tilting the angle over 5 degrees, the SHG increased significantly. This was mainly the case for the two nearly-overlapping hole arrays with sharp apexes, with center-to-center hole spacing of 190 nm and 195 nm. For all the other arrays, the SHG signal remained weak below 10° angle of incidence. For the 8° angle of incidence, there was a significant enhancement of SHG for the array with a 195 nm center-to-center distance between the two holes, whereas the 190 nm array showed maximum SHG for other angles.

For angles larger than 12°, the SHG decreased. This may be the result because there was no longer good phase-matching between the transmission resonance and the fundamental wavelength. The linear transmission of the fundamental wavelength also decreased as the angle of incidence was increased, which supports this hypothesis.

The maximum of SHG for each array arises for different angles of incidence; the larger the center-to-center distance, the smaller the maximum SHG angle of incidence. The main peak of the Bragg resonance is red-shifted by changing the angle of incidence [1]. This influences the resonant enhancement of both the pump beam and the SHG. Therefore, by changing the angle of incidence, the maximum local field enhancement can be varied.

By comparing the SHG maximum measured for each array, we calculated a 14 times ratio between the maximum for the array with a 0 nm (circular holes) and 195 nm spacing (double-holes). This significant ratio is explained by the local enhancement of the electric field at the nanoscale, close to the apexes. Although the SHG is enhanced by a factor of 14, the local field is expected to be enhanced by a significantly greater value since the SHG is coming from a much more localized area around the apexes. To explore this effect, we performed finite-difference time-domain (FDTD) simulations of double-hole structures to investigate how the proximity and sharpness of the apexes influence the local electric field intensity already presented elsewhere [10]. The electric field intensity calculated was four times higher for the double-hole structure with apexes than for a double-hole structure without apexes (oblate hole).

4. Conclusion

We have presented the first angle-dependent measurements of SHG from the double-hole structure. The double-hole structure in a gold film enhances the SHG when the holes are just overlapping to produce two nearly touching sharp apexes. This is due to the local enhancement of the electric field at the apexes. For different center-to-center hole spacings, the maximum SHG signal occurred for different angles. This effect came from the need to break the symmetry for SHG as well as from the shift in the Bragg resonances of the array with tilts to the sample.

Future work consists of varying the periodicity of the double-hole arrays to study its dependence. This should also permit to perform surface-enhanced Raman scattering measurements in order to show the potential of the double-hole structure for molecule detection.

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Figure 1: Scanning electron microscope image of double-hole array with hole-spacing of 195 nm. y and x polarizations are shown with arrows.

Figure 2: Schematic of SHG measurement setup in transmission geometry.
Figure 3: Transmission spectra for different hole-spacings (0 nm, 195 nm and 250nm)

Figure 4: Angle-dependent SHG measurements in transmission for different double-hole center-to-center hole-spacing