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Increase of group delay and nonlinear effects with hole shape in subwavelength hole arrays

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Abstract. We investigate the influence of hole shape on the group delay of femtosecond laser pulses propagating through arrays of rectangular subwavelength holes in metal films. We find a pronounced dependence of the group delay on the aspect ratio of the holes in the arrays. The maximum group delay occurs near the cut-off frequency of the holes. These experimental results are found to be in good agreement with calculations. The slow propagation of light through the array gives rise to enhancement of the second harmonic generated in the structures. The observed behavior is consistent with the presence of a resonance at the cut-off frequency of the rectangular holes.

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1. Introduction

Ever since its discovery by Ebbesen *et al* [1], extraordinary transmission (EOT) of light through arrays of subwavelength holes has been an intensively studied subject [2]–[4]. Previous research on the physics behind the effect of high transmission through such arrays has contributed to many important developments in nano-optics [5]–[7]. Also, EOT has led to the use of surface plasmon polaritons in ingenious optical designs, for instance to realize light beaming from a subwavelength aperture [8], and in novel spectroscopic devices [9].

An intriguing aspect of EOT is the large influence of hole shape [10, 11] on the transmission of light through rectangular holes [12, 13]. Changing the aspect ratio of a rectangular hole considerably redshifts the transmission peak of light polarized parallel to the short axis of the hole. Also the total transmission, normalized to the open fraction, increases for holes with larger aspect ratio. Recent theoretical work suggests that close to the cut-off frequency of a single rectangular hole, a Fabry–Perot-like resonance exists [14]–[16]. The importance of this resonance to the transmission of hole arrays was pointed out by Garcia-Vidal and co-workers [17]. The resonance occurs because at cut-off the propagation constant in the hole becomes very small. Thus, reflections within the hole add up constructively, leading to a Fabry–Perot-like resonance.

Most studies of hole arrays have been performed by analysing the spectrum of the transmitted or reflected light. Time-domain measurements can be used to obtain information complementary to these frequency-domain methods. The propagation of light pulses through subwavelength holes has been studied to investigate the group velocity of light propagation through these structures [18]–[21], showing for instance large negative delays. The role of hole shape in the propagation of light pulses through hole arrays has been investigated in the THz regime [22]. To the best of our knowledge, the role of hole shape at optical frequencies is yet to be explored.

The high transmission occurring in EOT leads to high electromagnetic fields in the holes. This has led to research interest in hole arrays for enhancing nonlinear effects [23] and sensing applications [24]. An investigation of the role of the aspect ratio of a rectangular hole in the second harmonic generation (SHG) efficiency of hole arrays [25] showed that a specific aspect ratio exists for which the SHG efficiency is an order of magnitude higher than for other rectangular hole shapes. It was found that this enhancement could not be explained by the linear transmission properties at the fundamental wavelength. Recent investigations of nonlinear effects occurring in photonic crystal structures have shown the important role played by the group velocity (v_g) in the efficiency of nonlinear processes [26, 27]. In the case of a second order nonlinear process such as SHG, we expect that the efficiency will scale as v_g^{-2} .

In this paper, we investigate the connection between the nonlinear effects and the pulse propagation through hole arrays. The group delay of a femtosecond pulse through an array of subwavelength holes is measured for different aspect ratios of the holes. Additionally, the amount of second harmonic generated by the same femtosecond pulses in the array is determined. We observe an increased group delay and concomitantly larger nonlinear effects near the cut-off frequency of a rectangular hole.

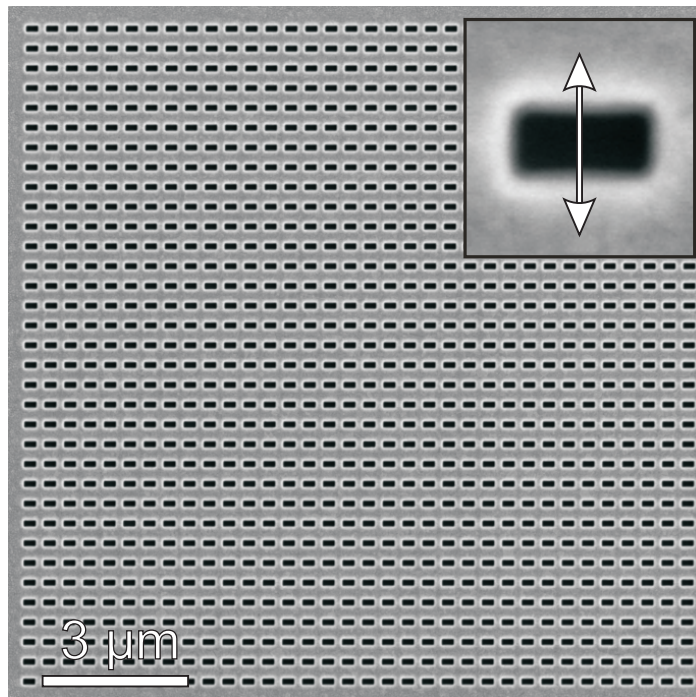


Figure 1. Scanning electron microscopy image of a hole array. The inset shows a close-up of an individual hole, in this case with sides $W = 246$ nm and $H = 117$ nm. The aspect ratio W/H is 2.1. In the experiments the polarization is oriented along the short axis of the holes, as indicated by the white arrow.

2. Experimental

The samples under investigation are arrays of 34×34 holes milled with a focused ion beam (FIB) in a 200 nm thick gold layer on a glass substrate (figure 1). Each structure is milled with the same conditions of the FIB, which should lead to a comparable surface roughness in each structure. The period of the square lattice array is fixed at 410 nm. The dimensions of the holes vary from 205×141 to 328×88 nm. The aspect ratios of the holes range from 1.46 to 3.73. This range is chosen so that for wavelengths around 800 nm the holes with the smaller aspect ratio are in cut-off, while the holes with larger aspect ratio are not [28]. The total open surface of the arrays is kept constant at $33 \mu\text{m}^2$. For calibration purposes every hole array is accompanied by a large reference hole with the same outer dimensions as the whole array, i.e. $14 \times 14 \mu\text{m}$. The structures and reference holes are placed $70 \mu\text{m}$ apart.

To determine the group delay we use an interferometric technique. Light from a 80 MHz Ti:sapphire oscillator (Spectra Physics Tsunami) is sent into a Mach–Zehnder interferometer (see figure 2). The center wavelength of the oscillator is tunable from 760 to 830 nm and the pulse duration is ~ 100 fs. The sample branch of the interferometer contains the sample, and the reference branch contains two acousto-optic modulators and a delay line. The light of both branches is combined with a fiber coupler and the interferometric signal is measured with an Si photodiode. In the sample branch the light is focused on the sample and collected using two lenses with a numerical aperture of 0.4. The focus is determined to be smaller than $2 \mu\text{m}$ by imaging the sample and the focus on a camera. To compensate for the dispersion

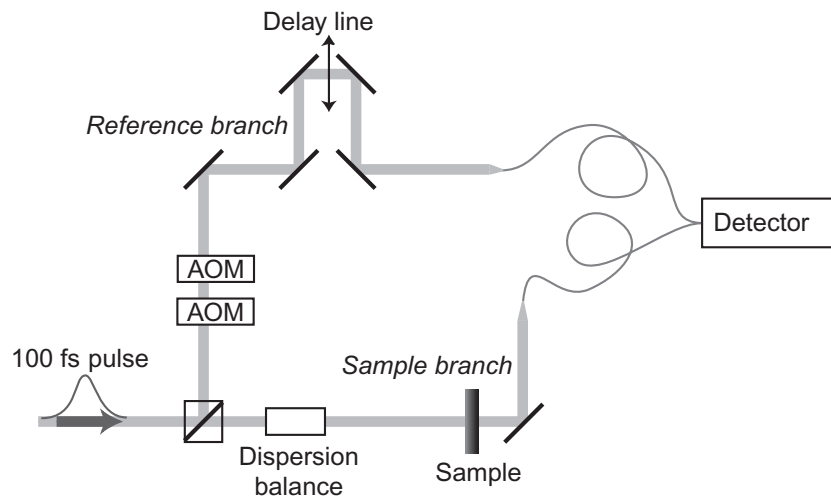


Figure 2. The interferometric set-up used to determine the group delay. Femtosecond pulses from a Ti:sapphire laser are split into the two branches of a Mach–Zehnder interferometer. In the reference branch two acousto-optic modulators are placed to shift the frequency of the light by 9 MHz. A delay line is used to measure the interferogram. In the sample branch a sample is placed, plus a dispersion compensating crystal to balance the dispersion introduced by the two acousto-optic modulators in the reference branch. Both signals are coupled into a 2-to-1 fiber coupler.

of the two acousto-optic modulators that are mounted in the reference branch, a dispersion balancing crystal is mounted in the sample branch. The two acousto-optic modulators in the reference branch shift the frequency in the reference branch by 9 MHz. This leads to a temporal 9 MHz beat signal in the detected interferometric signal. With a lock-in technique we obtain the amplitude and phase of the interference signal simultaneously. By moving the delay line an interferogram is measured as a function of the time delay. We compare interferograms through a structure to interferograms through a reference hole to obtain what we will refer to as a relative group delay. The real group delay can be derived from this relative group delay by simply adding the well-known delay of a pulse traveling through a layer of air with the same thickness as the gold film. The delay line used is a pair of mirrors mounted on a linear motor (Newport XMS50) with a linear encoder (Heidenhain LIF 481). The stage is moved at a constant velocity and a single interferogram is acquired in half a second. The polarization of the light in the experiment is rotated with a $\lambda/2$ wave plate to orient it parallel to the short axis of the holes. The sample is mounted on an X-Y piezo arm that moves the sample perpendicularly to the impinging laser beam. We collect 100 pairs of interferograms by quickly alternating between measurements on the hole array and on the reference hole. This procedure renders the method insensitive to long-term drift in the optical path lengths of the set-up. All the measured interferograms are numerically filtered in the frequency domain to remove low-frequency noise sources of electronic origin. To determine the group delay the light experiences when traveling through the structure, a Gaussian is fitted to the amplitude of each filtered interferogram. From the positions of the fitted Gaussians, an average group delay is determined, as well as a spread in measured delays.

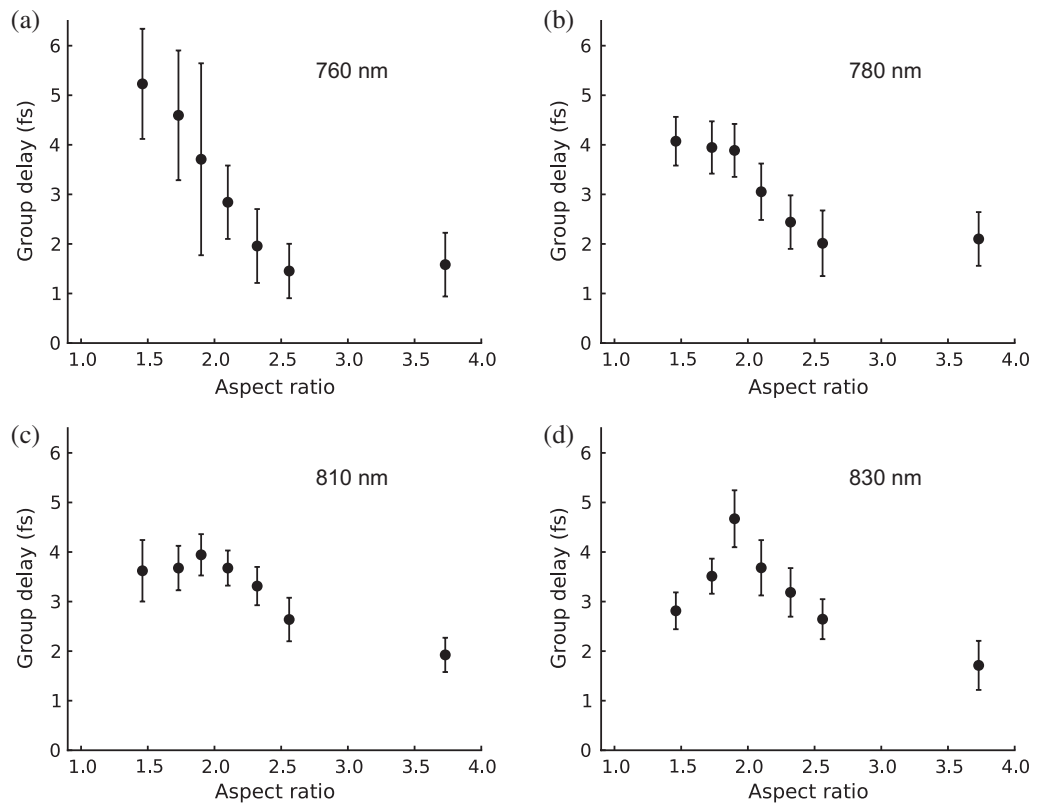


Figure 3. The group delay measured for hole arrays with different aspect ratio holes for a laser wavelength centered around (a) 760 nm, (b) 780 nm, (c) 810 nm and (d) 830 nm. The error bars are the standard deviation over 100 measurements. For reference, a pulse traveling through a reference hole would arrive with a delay of 0.66 fs in this figure.

The second harmonic generated in a sample is measured in a separate experiment. The light is focused on the sample with an NA of 0.17 and collected with a higher NA. The sample is tilted under a small angle of 2.5° to prevent back reflections from reaching the mode-locked pulsed laser. We verified in a separate experiment that the results depend only weakly on this angle. The transmitted fundamental wavelength is attenuated by a combination of colored glass and interferometric filters. The generated light is detected with a spectrometer (Acton SpectraPro 2300i) equipped with a cooled CCD camera (Princeton Instruments Spec10-B/XTE). Spectra are typically collected in 200 s. We verify that the signal is the second harmonic by checking that the signal at the doubled frequency depends quadratically on the input power.

3. Experimental results for the measured pulse delay

In figure 3 the group delay is shown as a function of aspect ratio for four different wavelengths. For all wavelengths and aspect ratios a positive delay is found with respect to transmission through a reference hole. The observed group delays, caused by the propagation of the pulse through the hole arrays, range from 1.5 to 5.2 fs. This is large compared to the 0.66 fs delay of a

pulse propagating through the same thickness (200 nm) of air. Figures 3(a) and (b) for 760 and 780 nm illumination both show a decreasing delay as the aspect ratio increases. The delay for pulses of 810 nm (figure 3(c)) shows a weak maximum near aspect ratio 2. Figure 3(d) shows the results for pulses with a center wavelength of 830 nm. Here a clear peak can be observed at aspect ratio 2.

The maximum delay τ_{\max} we find in our measurements is 5.2 fs. Thus the observed delay corresponds to a slow propagation velocity of the pulse through the 200 nm thick array of $c/8$. By simply altering the shape of the rectangular holes, the group delay through the hole array can therefore be tuned by a factor of three.

4. Modeling calculations of the group delay

We used finite integration technique (FIT) calculations [29] to complement our experimental findings regarding the group delay. The transmission of an ultrashort femtosecond pulse through an array of subwavelength holes was calculated for various aspect ratios of the holes. To calculate the group delay efficiently, periodic boundary conditions were used in the two in-plane directions. Thus the results of the calculation correspond to the transmission through an infinitely large array. Note that in the experiment a focused beam is used while the calculation is based on a plane wave. The spread in wavevectors caused by the focusing in the experiment thus leads to a slight averaging effect in the observed delay that is not included in the calculation. The dielectric constant of gold in this calculation was described with a Drude model, the parameters of which were obtained by fitting to measured values of the dielectric constant. The calculated fields before and after the structure, respectively $E_{\text{in}}(t)$ and $E_{\text{out}}(t)$, are Fourier transformed and used to determine the complex transfer function of the structures via $T(\omega) = \mathcal{F}[E_{\text{out}}]/\mathcal{F}[E_{\text{in}}]$. From this the group delay is determined using $\tau_g = \partial \arg[T(\omega)]/\partial \omega$.

When determining the group delay from either measured or calculated data, one has to be careful when spectral filtering takes place. The reason for this caution is that a combination of group velocity dispersion and spectral filtering can cause a delay of the pulse envelope in addition to the delay caused by a reduced group velocity. We can estimate the magnitude of this effect on the observed delay, using the results obtained in the FIT calculation. We determine the delay of a 100 fs Fourier-limited pulse $E_{\text{in}}(\omega)$ using the transfer function $T(\omega)$ and the normalized transfer function $N(\omega) = T(\omega)/|T(\omega)|$ that excludes the effect of spectral filtering. We calculate the delay of the output pulse shapes $E_{\text{out}}(t) = \mathcal{F}^{-1}[E_{\text{in}}(\omega)T(\omega)]$ and $E_{\text{norm}}(t) = \mathcal{F}^{-1}[E_{\text{in}}(\omega)N(\omega)]$ and find that the difference in delay is always smaller than 150, as for our experimental conditions, which is well within the experimental error. We therefore consider the effects of spectral filtering to be minimal in this system.

The result of these calculations is shown in figure 4, where the calculated group delays for arrays of holes is plotted for several wavelengths as a function of aspect ratio. The negative delays in the calculation are caused by the anomalous dispersion in the transfer function of the hole array and have been experimentally observed in previous work [19]. In the calculation, we see that the group delay exhibits a maximum, which shifts to higher aspect ratios as the wavelength is increased. This is very well reproduced by the measurements. For the 760 nm data (figure 3(a)), we see a monotonic decrease of the group delay as a function of aspect ratio. In figure 4, we see that this is because the peak actually lies at an aspect ratio smaller than that used in our measurements. It can also be seen from figure 4 that for wavelengths larger than 780 nm, the maximum in group delay lies in our window of aspect ratios. That is why in figures

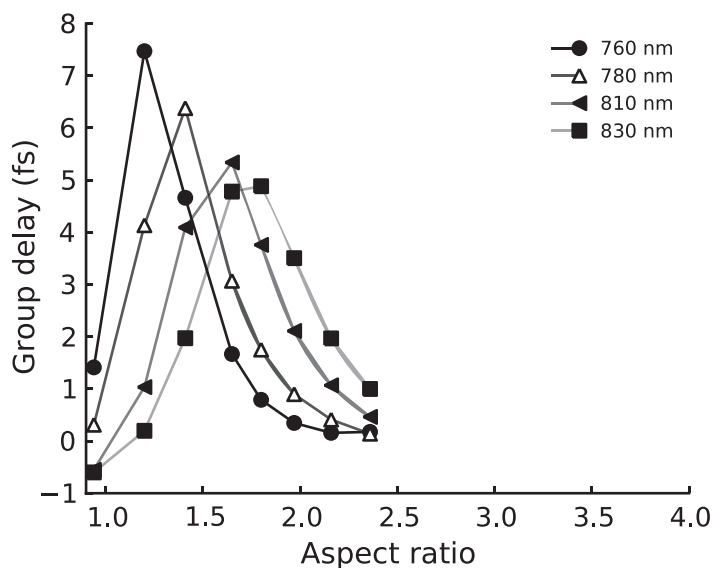


Figure 4. The group delay, calculated with a FIT, as a function of the aspect ratio for four different wavelengths: 760, 780, 810 and 830 nm.

(b)–(d) we indeed observe a maximum in group delay. There are, however, some discrepancies between the calculation and the experiment. For instance, the maximum group delay that is found in the experiment is roughly 20 per cent smaller than in the calculation. Furthermore, the maxima in group delay for 810 and 830 nm appear at a larger aspect ratio in the experiment (approximately 2) than in the calculation (approximately 1.7). We attribute these differences to a combination of uncertainty in the experimental determination of the geometry of the holes and the difference between the optical properties of gold used in the calculation—which for technical reasons is described by a Drude model—and the actual optical properties of the gold film.

5. Experimental results on the second harmonic generation (SHG)

Figures 5(a)–(c) show the SHG signal normalized to the incoming fundamental power squared as a function of the group delay that was observed. Due to the low detection efficiency of the Silicon CCD at 380 nm, no results were obtained for the measurements of the SHG with 760 nm fundamental wavelength. In each graph, the order of magnitude of the SHG per Watt squared is comparable. Most strikingly, for the other three wavelengths, a strong increase of the SHG signal with group delay is observed. From the observed trend in the SHG as a function of observed group delay, we infer that the resonance leading to an increase in group delay also leads to an increase in nonlinear effects. The data are consistent with quadratic dependence on group delay. Unfortunately, the data quality does not allow us to reliably fit a power law. A few data points deviate from the trend observed between SHG and group velocity, most notably the point with the highest SHG count per squared Watt in figure 5(c). This suggests that besides group velocity, additional factors play a role in the generation of the second harmonic.

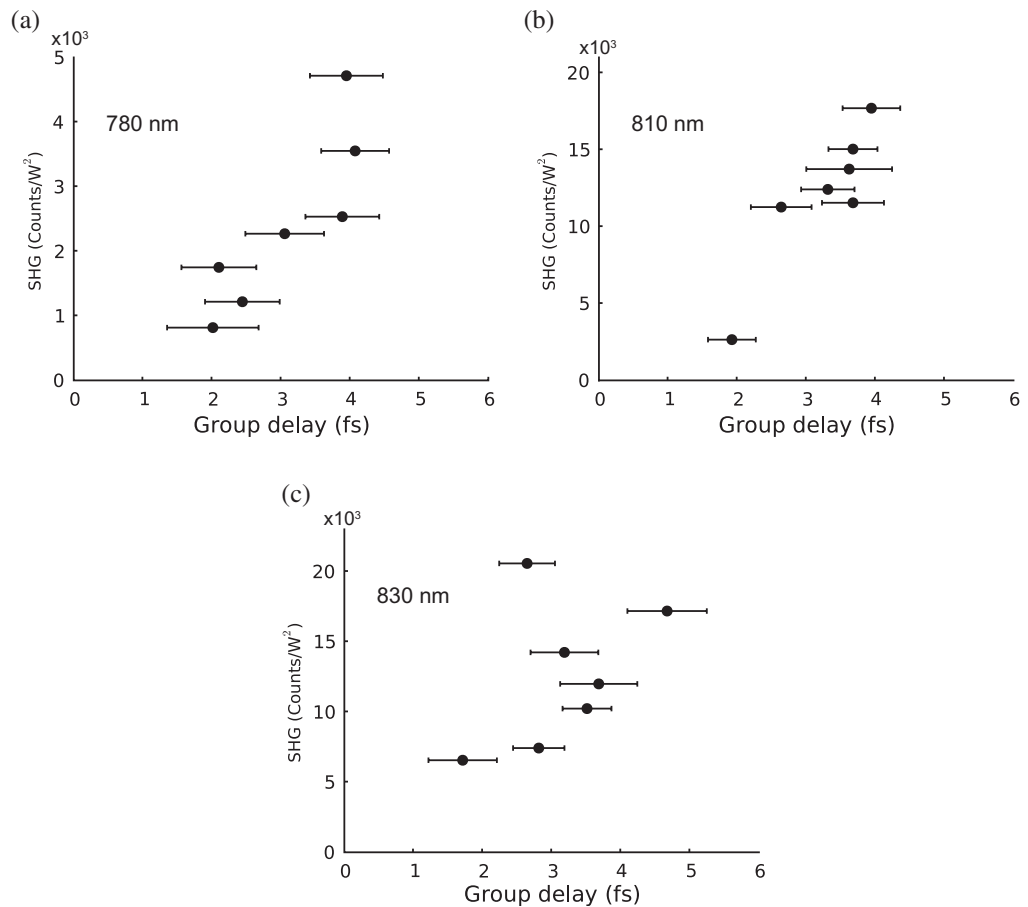


Figure 5. The SHG versus the observed group delay for three different wavelengths: (a) 780 nm, (b) 810 nm and (c) 830 nm. All three graphs show that there is an increase of the SHG signal as the group delay increases.

6. Discussion and conclusion

We investigated the influence of hole shape on the group delay of femtosecond pulses propagating through hole arrays. We observed a maximum delay that shifts to larger wavelengths for holes with larger aspect ratio. This is in agreement with finite integration calculations and theoretical work that predicts the presence of a resonance at the cut-off frequency of subwavelength holes [17]. Measurements of the light generated by SHG in hole arrays show a clear correlation with the enhanced group delay present at the cut-off resonance. This suggests that the enhanced group delay gives rise to larger nonlinear effects. We have to consider, however, that the observed correlation between SHG and group delay does not prove a unique causal relation between both. In fact, we anticipate that at least two additional factors play a role in this phenomenon. As the hole shape is varied, the linear transmission and the mode profile in the holes change for both the fundamental and the second harmonic frequency. Previous work [25] did, however, show that the linear transmission of the fundamental frequency cannot fully account for the observed enhancement of the SHG. The measurements in this paper indicate that the group delay is an important factor. Both experimentally and theoretically, it is

very difficult to identify the role that the group velocity, linear transmission and mode profile play. The results presented here are an important step forward as they show for the first time the striking correlation between the SHG and the group delay in these structures. To know the exact mechanism of the SHG in these structures, however, will require further study. Gaining deeper insights into the role of surface roughness is of particular importance.

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