Second harmonic generation enhancement in lithium niobate micro-tips

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Abstract: We have observed a substantial enhancement (40 fold) of the non-phase-matched second harmonic generation efficiency in microfabricated ultra-sharp, lithium niobate micro-tip arrays. The fabrication process and the nonlinear characterization is presented.

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1. Introduction

Research on methods for microstructuring lithium niobate has been attracting substantial interest lately as this would provide the means for expanding the utility of this very important nonlinear optical ferroelectric crystal. A wide range of microstructuring methods has been reported up to now which are based on differential etching in hydrofluoric (HF) acid induced by either ferroelectric domain inversion [1] or UV illumination [2], or other dry etching methods such as ion beam milling [3] and laser ablation [4].

Ferroelectric domain engineering in particular can be applied over a large area of a z-cut lithium niobate substrate (wafer) creating a regular or arbitrary distribution of inverted ferroelectric domains which can be subsequently converted into a high aspect ratio surface relief structure due to the polar-surface-specific etching in HF acid. In this way it is possible to fabricate micron-size 1D linear gratings or 2D grating arrays with very sharp features corresponding to the ferroelectric domain pattern. The resulting structures are of excellent optical quality and they are suitable for optical transmission experiments. In this work we are reporting on the 40-fold enhancement of the non-phase-matched nonlinear optical second harmonic generation efficiency that has been observed in single crystal micro-tips which have been fabricated by ferroelectric domain engineering followed by chemical (HF) etching.

2. Fabrication process

Regular arrays of micron size tips has been fabricated from a 0.5 mm thick z-cut congruent lithium niobate single crystal wafer (crystal technology Inc. USA). In the first step of the fabrication process arrays of hexagonally shaped inverted domains arranged in a regular hexagonal lattice was obtained by electric field poling at room temperature [5]. Two different sets of samples were fabricated having different periodicities where the distance between first neighbors in the unit cell of the hexagonal lattice could be either 39 µm or 16 µm respectively with good uniformity over the whole microstructured area which was of order 2 cm². In the second fabrication step the ferroelectric domain engineered sample was placed in a hot (500°-600°C) HF acid bath for several hours for the differential etching to occur. After approximately 15 hours of etching an array of pyramidal structures (corresponding to ±2 polar surfaces) with triangular cross-section was revealed as shown in the Scanning Electron Microscopy (SEM) picture in figure 1.

Fig. 1. Top SEM view of a regular array of pyramidal surface relief structures (tips) with a base width of 30 µm. The inset in the bottom left shows a high magnification side view of the ultra-sharp end point of an individual pyramid.
The width at the base of the particular micro-structured array shown in figure 1 is \( \sim 30 \, \mu m \) while the height is \( \sim 70 \, \mu m \). The inset figure shows a high magnification SEM side view of the ultra-sharp end point of an individual micro-pyramid. Although the individual inverted ferroelectric domains have a hexagonal shape the final etched structure has a triangular cross section which is due to an additional differential etching process which takes place along the three crystal symmetry directions on the x-y plane. However, differential etching of the inverted domains also takes place on the face opposite the one shown in fig. 1. In order to maintain the good optical quality of that face for optical experiments this face was covered with an HF resistant material during the etching process.

3. Experiments and discussions

The micro-tips used in the second harmonic generation experiments had a base diameter of \( \sim 16 \, \mu m \) and a height of \( \sim 70 \, \mu m \). The beam from a mode-locked titanium sapphire laser (Coherent MIRA oscillator coupled into a RegA amplifier) was coupled into individual tips after being focused by an f=45 mm plano-convex lens. The peak wavelength was 793 nm and temporal width of the pulses was 160 fs at a frequency of 100 kHz. The spot diameter of the beam at the focal point was calculated to be of order \( \sim 15 \, \mu m \) which is comparable to the base width of the individual tips. The average power range of the input laser beam was between 0.30 mW and 40 mW. The corresponding energy per pulse was between 3 nJ and 400 nJ, while the intensity range at the focal point was between 30 GW/cm² and 1.7 TW/cm². These values are corrected for the transmission losses of the lens and the reflection losses on the air-crystal interface. Self focusing and channelling effects, which limit the optical throughput of the structure were observed at higher input power levels. However, within the range of input power used in this experiment non-phase-matched second harmonic radiation could be observed in the optical output of individual tips.

The output from individual tips was subsequently collected by a microscope objective and measured with a power meter. The measured output power of the second harmonic radiation follows a quadratic power dependence with respect to the measured power output of the fundamental wave as shown in figure 2. This quadratic power dependence confirms that there were no nonlinear power dependent scattering processes taking place within the optical power range used in this study. Such nonlinear power dependent scattering processes were observed however at higher input optical power levels.

![Figure 2. Second harmonic output power as a function of the output of the fundamental. The solid line corresponds to a quadratic fitting function. Inset: light propagation inside a micro-pyramid.](image)

The efficiency of the nonlinear second harmonic generation in the microstructured tips was estimated by the ratio between the measured optical power of the second harmonic and the optical power output of the fundamental wave. Due to the geometry of the tips the fundamental wave which was originally directed along the z direction of the crystal undergoes total internal reflections on the side surfaces of the pyramidal structure changing its direction, as shown in the inset of figure 2. Subsequently, other nonlinear coefficients (including the large \( d_{33} \) coefficient) may participate in the nonlinear process hence it is important to compare the conversion efficiency observed in the microstructured tips with the corresponding efficiency for unstructured samples of the same thickness but of different crystal cut where the \( d_{33} \) coefficient can be accessed.

Figure 3 shows the measured percentage conversion efficiency as a function of the average optical power output of the fundamental wave for the three different cases of: i) a single microstructured tip, ii) unstructured x-cut, and iii) unstructured z-cut samples. The SHG conversion efficiency in the case of the z-cut microstructured tips is three orders of magnitude higher as compared to the recorded efficiency of the unstructured z-cut samples. The choice of the x-cut sample was in order to utilize the highest \( d_{33} \) nonlinear coefficient.
The second harmonic generation for unstructured \( z \)-cut samples was very low and it was necessary to increase substantially the input power in order to get an appreciable signal. The high intensities used in this case lead to the reduction of the conversion efficiency observed at higher power values of the fundamental wave due to parasitic nonlinear effects (self focusing, channeling) as shown in figure 3. However, the efficiency of the microstructured sample appears to be far higher than that of the \( x \)-cut sample as it corresponds to lower optical power of the fundamental wave.

![Graph showing second harmonic conversion efficiency](image)

**Figure 3.** Plot of the % second harmonic conversion efficiency for the microstructured tips (triangles) unstructured \( z \)-cut (circles) and unstructured \( x \)-cut (squares)

The output power of the fundamental wave corresponding to equal values of the conversion efficiency was up to ~400 times lower in the case of the tips as compared to the output power of the \( x \)-cut sample. However, the confinement of the fundamental wave in the tip due to total internal reflections should increase the local intensity. The area of the tip where the second harmonic is generated (estimated by imaging the output area of a single tip with a CCD camera) is 10 times smaller with respect to the area at the base of the tip resulting in a 10 fold increase of the optical intensity. Assuming that the \( d_{33} \) nonlinear coefficient is fully utilized in the case of the tips (which is an overestimation because of limiting geometrical and polarization parameters) the measured conversion efficiency in the case of the tips appears to be 40 times higher than the expected one.

Since there is no phase-matching condition satisfied in the nonlinear SHG process the only parameter that could be responsible for the efficiency enhancement is the actual optical nonlinearity of the material. The optical nonlinearity is strictly related to the configuration of ions in the crystal lattice and consequently our results suggest that this ion configuration is modified in the vicinity of the tip. The ion configuration in the crystal may be affected for example by the strong electric field which is associated with the presence of the ultra-sharp single crystal tip in this ferroelectric material. Furthermore, electrostatic force microscopy measurements which are currently under way would evaluate the electric field enhancement in the vicinity of the tip thus, providing further insight into the observed effect.

4. Conclusions

Nonlinear SHG experiments in lithium niobate surface microstructures have shown a significant increase of the efficiency of the nonlinear process as compared to the efficiency of the same process in unstructured samples. The observed enhancement of the nonlinear SHG is attributed to the enhancement of the optical nonlinearity associated with sharp crystal edges. This observation implies the attractive prospect for more efficient micro nonlinear optical devices.

5. References