1 Introduction

In the wave optic theory of light, frequency is considered immutable, unlike wavelength which is easily altered by passage in a transparent material. Nevertheless, methods do exist for changing the frequency of light, and these turn out to have immense practical significance.

The equation $v = f\lambda$ describes the relation between the velocity $v$, frequency $f$, and wavelength $\lambda$ of light. In a vacuum, the velocity of light is $c$, a universal constant of nature. In any other transparent medium, light travels at a lower speed $v = c/n$, which depends on $n$, the index of refraction of the medium. The reduced velocity of light causes a proportional shortening of the wavelength, but in this process, the frequency remains unchanged.

There are many important reasons why one would want to change the frequency of light. As one example, it is sometimes necessary to change the frequency of light because the available lasers today cannot produce light of all different wavelengths. For especially specific wavelength demands, such as the excitation of an atom for laser cooling and other physics experiments, the frequency of an existing light source has to be altered to precisely match the frequency of light at which the atom operates.

Of the several possible ways of changing the frequency of light, I previously studied the relatively small frequency shifts created as a result of interactions between sound waves and light waves in an acousto-optic modulator (AOM). I also built a modest Michaelson interferometer to further research frequency alterations and to witness the Doppler shift created by the movement of one of the two mirrors. Finally, I researched a phenomenon found in optical fibers known as Stimulated Brillouin Scattering which also produces frequency shifted light with sound and light waves.

Nonlinear interactions between light and materials offer the possibility of a much more dramatic variation of frequency, such as doubling or even tripling. A nonlinear crystal is used to frequency double light because atomic interactions within the crystal create a polarization component at twice the frequency of the input light. Frequency doubling of laser light was
first demonstrated in 1961 but to this day remains technically very challenging. In contrast, linear crystals cannot be used because the input wavelength, speed and frequency are the same as that of the output.

There are many methods used to make multiple passes through a nonlinear crystal, but many complications arise from the procedures. One major problem is that the light is hard to manipulate under those circumstances. These complications can be circumvented with the use of a Periodically Poled (PP) nonlinear crystal as demonstrated in the experiments described in this report.

2 Applications of Frequency Doubling

Generation of coherent radiation by means of frequency doubling – especially into the blue and UV spectral regions – has attracted much interest. Possible applications of doubled blue and/or UV light include advancements in high-density optical data storage which creates a four to eight-fold increase in the amount of information a compact disk could retain.

Instead of laser-diode pumped solid state lasers, which work best from remote sensing stations but cannot generate the requisite ultraviolet wavelengths, frequency doubled light could be used. NASA is currently researching this technology to create a UV light source that would monitor ozone levels in the atmosphere from space stations. Frequency doubling of three wavelengths to produce a red-green-blue image projector can improve the field of entertainment by creating a large laser projection display [1].

Frequency doubled light could also be utilized by the field of biotechnology as an economical and compact way to perform fluorometry, cytometry and confocal microscopy as opposed to the bulky and expensive Argon lasers used otherwise. Recently, fluorometry has been used to separate sperm cells that would produce a female child and those which would produce a male child because that of the female fluoresces under the blue light about 3% more than that of the male [2].
3 Periodically Poled Structures

Although frequency doubling by means of nonlinear crystals had been theorized and performed since the invention of diode lasers in the 1960s [3], the periodically-poled (PP) crystal did not come about until the early 1990s. The production of a PP crystal by means of an electric field poling technique was first used on LiNbO$_3$ by Yamada et al. in 1993 [4]. Although the PP technology is by now fairly well known in optics laboratories, it has not yet been commercially perfected and PP crystals cannot simply be purchased on the market.

3.1 Theory

In the traditional methods of frequency doubling in a nonlinear crystal, the achievable conversion ratios are subject to significant inherent limitations related to the length of the crystal in comparison to the wavelength of light. Although the crystal should be as long as possible to maximize the frequency-doubled light, if it is too long, light production will be limited by phase mismatch caused by the dispersion of the second harmonic light in the crystal.

There are tricks that could be performed to avoid these complications. A possible solution would be to calculate precisely the length at which the second harmonic would interfere destructively with itself in the specific index of refraction of the crystal and to cut the crystal accordingly. Even though the resulting crystal would be relatively short, it could be placed in a buildup cavity to boost the output of the generated light through multiple passes [5]. However, there are drawbacks to having a cavity in that if surface losses result from reflections, the doubling efficiency would be greatly degraded. Often, to reduce surface losses, the crystal is oriented for the suitable polarization and is also cut at Brewster’s angle.

All these problems with dispersion, phase mismatch, surface losses and orientation of the crystal can be eliminated with the use of a periodically poled crystal. As suggested by its name, a periodically poled crystal is a row of orientation-altered microscopic crystal slices of a nonlinear crystalline material. This technique eliminates the phase mismatch previously
encountered because before the light can disperse, creating a significant phase discrepancy, it is forced to turn as it enters into the next slice (Figure 1).

The total power of the second harmonic generated in a PP crystal can be described as

\[ P_{\text{Total SHG}} \propto \left[ \sum \frac{E_i^2}{A_i} \right]^2 \]

The power of the second harmonic is proportional to the sum of the electric field squared of the fundamental beam \((E^2)\) over the illuminated area of each poled slice of nonlinear material \((A)\), squared. The equation implies that the second harmonic is proportional to the total length squared, suggesting that the crystal can be arbitrarily long, producing an large amount of second harmonic. This occurs provided that the laser beam can remain small throughout the length of the crystal and that the primary laser beam does not get depleted.

### 3.2 Suitable Materials: Lithium Niobate (LiNbO₃)

There are many nonlinear materials that can be used for frequency doubling: KTiOPO₄, LiIO₃, LiTaO₃, KDP, KnbO₃ and many others. Each crystal has its strengths and weaknesses, but LiNbO₃ is often the preferred choice. Lithium niobate is a man-made material that has a high effective nonlinearity coefficient \((d_{33} = 27 \text{ pm/V})\) and is transparent over a wide light spectrum, from 0.35 \(\mu\text{m}\) to > 5 \(\mu\text{m}\) [6]. Because of its properties, it is considered to be the most suitable material for second harmonic generation of infrared light. Chemically, it is composed of planar oxygen sheets with lithium and niobium atoms lodged off-center between these planes. The optical nonlinearity found within lithium niobate can be explained by the asymmetry in the ferroelectric crystal structure [7].

### 3.3 Construction

The natural orientation of the polarization in LiNbO₃ can be reversed by applying a strong external electric field along the crystalline \(z\)-axis, with the field direction opposite to that of the original domain polarization. This electric field moves the niobium ions between the
oxygen planes and the lithium ions to the opposite side of the planes. This process is what is known as poling as the name derives from the creation of antipodes.

Unfortunately, the high room temperature coercive field required to pole lithium niobate is close to and sometimes overlaps the dielectric breakdown field of the crystal [8]. Furthermore, a pyroelectric material such as LiNbO₃ can develop a buildup of opposite charges which, if large enough, could create domain inversion.

Nevertheless, periodic poling can be achieved by photolithographically patterning electrodes on the exterior of a LiNbO₃ crystal in a lattice configuration. After the photoresist grating pattern is printed on the crystal, a liquid electrolyte makes electrical contact to the notched areas in the photoresist grating. Finally, the endfaces of the successfully poled crystal are polished to insure high transparency [9].

4 Experimental Arrangements

In the following section, the various components of the experimental setup and their arrangement, and the experimental procedures that were followed will be described.

4.1 Components

The main components of the setup are the PPLN crystal, the diode laser assembly used as the emitting light source and its electronic controller, the optics used to focus, collimate and filter the laser beams, the heater for the crystal, and the photodiodes that detected the 1310 nm and second harmonic light. A minor but essential component was an infrared viewing card obtained from Radio Shack for $5. This was in constant use for monitoring and adjusting the path of the 1310 nm laser beam.

4.1.1 My Periodically Poled LiNbO₃ Crystal (PPLN)

The sample periodically-poled lithium niobate crystal for my experiments was kindly provided by Dr. Leo Hollberg of the National Institute of Standards and Technology (NIST) in
Boulder, Colorado (Figure 1). The crystal has 4 different poling periods ranging from 12.4 \( \mu m \) to 12.7 \( \mu m \) in four separate tracks, each 1-2 mm wide.

![Diagram of PPLN crystal with poling periods](image)

Figure 1: PPLN crystal. The crystal is 18 mm in length along the direction of the light beam and 9 mm wide, but only 0.4 mm high.

The poling periods mentioned above are designed to match light with \( \lambda \approx 1310 \) nm, but since the laser wavelength is not precisely fixed in advance or easily tunable, the crystal itself must be ‘fine tuned’ by varying its temperature, as described in 4.3.2.

### 4.1.2 1310 nm Diode Laser

A 6 mW 1310 nm InGaAsP diode laser was obtained from ThorLabs for $265 to suit the needs of the crystal. A diode laser can be tuned by precisely changing the temperature of the laser [10]. Because the equipment to do so was not available, the crystal must be heated so that the crystal can tune into the exact frequency emitted by the laser.

A ThorLabs Laser Diode Controller was set to provide a constant input current of 22.5 mA. Since the laser beam is highly linearly polarized, it is vital that the input laser polarization is perpendicular to the large surface of the crystal while the beam itself propagates normal to the small surface.
4.1.3 Optics

Probably the most significant experimental challenge of this experiment was to properly match and focus the collimated light emitted from the diode laser assembly to the tiny proportions of each track in the crystal. This task is roughly equivalent to fitting a flashlight beam through a drinking straw from a distance of several meters, with the help of a magnifying glass. In this case, the ‘magnifying glass’ was a simple single-element lens, part of a lens kit from ThorLabs, with a 1 inch diameter and a focal length of 250 mm. The equations and the methods used to select this particular lens and calculate its focal properties are discussed in Section 5, Gaussian Optics.

Another important optical element was an interference filter (03 FIB 014) from Melles Griot placed right after the PPLN crystal to block 1310 nm light from the 655 nm light detector. This filter blocked out more than 99.9% of the infrared light and allowed more than 70% of the doubled light through.

For the first observations of 655 nm light, before the interference filter was received, a prism was used to separate the two types of light. The result was two distinct beams ~3 mm apart, one visible and the other visible only with an infrared view card.

4.1.4 Crystal Holder

The crystal holder has to do several things. It has to securely hold the tiny crystal without squeezing it, it has to help align the crystal to the laser beam, and it has to be able to heat up the crystal to as much as 80 C to achieve a matched frequency condition. The crystal holder was machined out of an aluminum block as shown in Figure 3. Its top surface has a shallow pocket just slightly (less than one mm) larger than the crystal. There are two large holes for two 50 Ω 2 watt resistors, which act as the heaters, and a small hole closer to the crystal for monitoring the temperature with a thermocouple.

The crystal holder is mounted on a three-axis micrometer translator that allows motion
of up to 10 mm in steps of only 10 microns in the $x$, $y$ or $z$ directions. A sheet of plexiglass ~0.5 inch thick was placed in between the crystal holder block and the translator to reduce heat loss by conduction.

**Figure 2. Crystal Holder**

### 4.1.5 Detectors

Because frequency doubling produces two very different frequencies, two types of detectors with very different response curves were required. A 0.2 mm$^2$ Germanium detector (ThorLabs DET 310) that measures 0.6 Amps per Watt at 1310 nm was used for the IR light. The second harmonic was detected by a 13 mm$^2$ silicon detector (ThorLabs DET 110) with a sensitivity of 0.45 Amps per Watt.

It was found that although the Ge detector had no response to red light placed in its path, the Si detector would register 0.2 mV when placed in the 1310 nm beam. It is believed
that the 1310 nm light heats the photodetector enough to somehow create a small voltage signal. Because of this finding, the interference filter was used to block out most of the 1310 nm light so that it would not affect the readings of the second harmonic light.

Voltmeters were used to numerically project the readings of the photodiode. A resistor of 1 kΩ was used to allow the fundamental light intensity to be displayed in Volts. As for the Si detector used for the frequency doubled light, the resistor was that of 1 megaΩ and the intensity of the second harmonic was measured in mV.

4.2 Layout

All of these components are mounted on an optical table for stability (Figure 3).

Figure 3. Experimental Setup

This configuration is only the basic setup used for most of my experiments and is changed slightly for various experiments. This setup has great potential for compact packaging.
4.3 Procedures

As previously stated, the setup altered as the experiments varied, and the procedures used also differ. Even so, the one constant in these procedures is that all of the final data were taken in track 3 in a completely darkened room with the voltmeter, thermocouple and a flashlight placed in a blackbox because the Si detector is sensitive to room light and measurements must be made in the absolute darkness. The blackbox is simply a cardboard box with its corners taped up and a garbage bag taped over it like a curtain to ensure that no light would be detected. Readings can be taken by taking cover under the garbage bag that is big enough to reach the floor, allowing for absolute darkness.

4.3.1 Alignment

Proper alignment is essential for producing second harmonic light because the spatial parameters within which the laser beam must maneuver are very limited. Any slight tilt may affect the way the Gaussian Beam propagates through the crystal and how much doubled light is produced.

The first part of the alignment procedures involved making the diode laser beam parallel to the surface of the table. Next, the focusing lens was introduced and centered on the beam axis. This alignment could be checked by making sure that the lens only focussed and did not deflect the beam.

The final and most difficult step was to align the crystal holder and the crystal such that the laser beam could entirely propagate through the crystal, parallel to the length of the track. The alignment was checked by scanning the laser through the gap available for the crystal vertically and horizontally to see how quickly the laser beam gets eclipsed by the edge of that gap. If the edge is tilted, the peak of the intensity graph picked up by the detector would be more narrower than the peak of the scan performed when the edge was perfectly parallel to the propagating laser beam.
This experiment was then repeated with the crystal inside the gap to monitor the transmission of 1310 nm light through the crystal and to find the place where optimal second harmonic generation may occur. The vertical scan was performed to find the best height where the most second harmonic generation is predicted. Where there appeared to be a peak in transmission of 1310 nm light, it was believed that it is there that the intensity of frequency doubled light would also be greatest (Figure 4). The horizontal scan of 1310 nm light was an attempt to measure precisely the width of the different tracks. The two high peaks at the ends of the graph represent the gap surrounding the crystal and the dips in the rather flat top signify the juncture between the tracks (Figure 5).

![Graphs showing intensity vs distance for vertical and horizontal scans](image)

**Fig. 4:** Vertical crystal intensity scan of 1310 nm light.  
**Fig. 5:** Horizontal crystal intensity scan of 1310 nm light.

### 4.3.2 Temperature Control

Temperature becomes crucial in this experiment because the crystal had to be manipulated by changing its temperature to conform to the frequency of light generated by the diode laser. In order find the best temperature at which the doubled frequency would be most intense, temperature scans were made. This was accomplished by turning the power supply to 15 volts and as the temperature increased, taking intensity readings of the 655 nm light in the dark in increments of 0.5 °C.
5 Gaussian Optics

The coherent radiation generated by a laser is typically a TEM$_{00}$ Gaussian mode that is most intense in the center and gradually decreases in intensity as the distance from the center of the beam increases. The beam itself propagates in a hyperbolic shape whose focus can be made steeper or shallower with the use of lenses. Although the Gaussian beam expands, it maintains the same Gaussian beam profile. A complete theory of second harmonic generation using Gaussian beams was first presented by Boyd and Kleinman in 1968 [11].

As noted before, the task of getting the laser light through the crystal is equivalent to trying to focus the light through a straw from a distance of several meters. Thus, the design of the optics strived to make the beam as small as possible within a track in the crystal.

![Diagram](image)

**Figure 6. Confocal Parameters**

Knowing that the length of the waist has to fit within 20 mm, the smallest waist possible on average was calculated with a Rayleigh Length ($Z_R$) of 10 mm. Working backwards,

$$w_0^2 = \left( \frac{\lambda Z_R}{\pi} \right) = \left( \frac{1.31 \times 10^{-6} \text{m} \times 0.01 \text{m}}{\pi} \right) = 4.17 \times 10^{-9} \text{ m}$$

Where $w_0$ is the radius of the Gaussian beam waist and $Z_R$ is half the length of the waist (Figure 6). It follows that $w_0 = 6.46 \times 10^{-5}$ or 64.6 $\mu$m.
The beam waist, $w_0$, is then substituted in the second equation
\[
Z = \left(\frac{\pi w_0^2}{\lambda}\right) = \left(\frac{\pi \times 6.46 \times 10^{-6} \times 511 \times w}{1.318 \times 10^{-6} \times \lambda}\right) = 154.92w
\]

Where $Z$ is the focal length of the lens to be used and $w$ is the radius of the input laser beam. Because the input laser beam radius ($w$) was measured to be about 1.75 mm, the focus length of the lens to be used is about 25 cm.

### 5.1 Beam Profile Measurements / Razor Blade

The Gaussian beam produced by the diode laser was profiled with the use of the 0.2 mm² Germanium photodetector which had a small enough aperture such that the detector could be used without the need for a pinhole. The photodetector was placed on a translator and measurements were taken in increments of 0.05 mm or 50 μm, scanning across the diameter of the laser beam as the intensity readings were recorded.

The intensity $I(r,z)$ of a Gaussian beam is described by the equation
\[
I(r,z) = \left[\frac{w_0}{w(z)}\right]^2 I_0 \exp\left[-2r^2/w(z)^2\right]
\]

where $z$ is the coordinate along the beam direction and $r$ is the radial coordinate perpendicular to the beam direction. The theoretical curve with the optimum width parameter of 1.75 mm (uniform line) is plotted on the same graph as the experimental measurements (symbols) and it can be seen that the two are almost identical (Figure 7).

### 5.2 Spreadsheet Model of the Experiment

A spreadsheet model of the Gaussian beam at the location of the crystal was created to see how well the beam would fit inside the crystal. The specifically predicted model was then tested by measuring beam sizes with the razor-blade method described by Suzaki and Tachibana [12]. As discussed in Section 7, Current and Future Work, this first version of the Gaussian beam spreadsheet model did not take into account that the crystal would modify the propagation of light.
The increasing radius of the beam waist, \( w(z) \), is described by the equation
\[
w(z) = \left[ (b^2 + 4z^2)/(2zb) \right]^{\frac{1}{2}}
\]
Here, \( \lambda = 1310 \) nm and \( b \) is the confocal parameter defined by Kogelnik and Li [13]. The confocal parameter is not a free parameter but is instead determined by the beam wavelength and waist size through the relation \( b = (2\pi/\lambda)w_0^2 \), where \( b = 2Z_R \).

6 Experimental Results and Discussion

Temperature scans were performed to measure the dependence of second harmonic light output on temperature. The optimum temperature for second harmonic generation in the third track was 33 degrees Celsius (Figure 9). Scans were also performed on the other three tracks, but no detectable second harmonic was produced.

The second harmonic intensity was graphed as the laser beam scanned across the width of track 3. Because the second harmonic generation graph had an irregular pattern, it had to be determined if these fluctuations were inherent in the crystal or was incidental. It was hypothesized that this occurred because the endfaces of the crystal were not polished uniformly. Two readings were taken as the laser beam entered from the left and after having flipped the crystal, two more were taken from the right to confirm accurate readings. The
measurements from the two sides were indeed different, yet they both had certain dips and peaks in common. An average was taken and is represented in the graph by the dark line.

Possible reasons for the fluctuations might be as proposed, but another rationale may be that the poling periods are not perfectly consistent or perfectly spaced with one another along the width of a track. Or, being that LiNbO₃ is highly susceptible to minute variations in the index of refraction, it may be a change in the index of refraction due to some unaccountable variable that generates this oscillating second harmonic generation.

Another experiment was executed to see if the output intensity was indeed the square of the input power times a constant. Neutral Density filters and microscope slides were placed between the laser and the focusing lens to reduce the power of the input light. The Germanium detector was first used to measure the attenuation of each filter and the slide and the many readings were averaged to find the amount of light transmitted by the medium.

Having taken the Germanium detector away, the second harmonic intensity was recorded at the optimal temperature, constant height and at 3.58 mm across where the peak second harmonic intensity reading existed on the horizontal scan (Figure 10).

The power of the input or output light is given by the equation \[ P = \left( \frac{I}{W} \right)^{2} \]. Since the

Fig. 9: Temperature dependence of Second Harmonic light in track 3. The three lines plotted are two successive scans and the difference between them.

Fig. 10: Horizontal scan of Second Harmonic light.
measured voltage $V = iR$ the power is determined by $P = \frac{V^2}{R(A/W)}$.

Figure 11 shows that the ratio of output power versus input power is indeed quadratic. The triangles are the experimental values, the solid line has the form $P_o = R(P_1)^2$, and the dotted line is to demonstrate that the experimental values do not have a linear relationship.

7 Current and Future Work

As mentioned in Section 5.2, the initial calculations of the Gaussian beam waist did not take into account the crystal’s properties, namely its index of refraction. After discussing this dilemma with a graduate student at Stony Brook, the following expression for the beam size $w(d_2)$ within the crystal was derived [14]:

$$w(d_2) = w_0^2 \left( \frac{d'^2}{z_0^2} + \left( \frac{d'}{f} - 1 \right)^2 \right)$$

Here $z_0$ is the Rayleigh length of the initial beam before the lens, given by $z_0 = \pi w_0^2/\lambda$, $d'$, is the effective distance $(d_1 + d_2)/n$, and $f$ is the focal length of the lens.
This correct expression has been incorporated into the new spreadsheet model which divides the crystal into 180 slices in order to mathematically predict the behavior of the light within the crystal. This spreadsheet can accurately project the size and shape of the beam waist in the crystal and the amount of total light generated. Recently, following the predictions, actual measurements of the second harmonic light were taken as the beam waist was manipulated by means of lenses with different focal lengths. Following the model, the crystal was placed, taking in account the refractive properties of the crystal, such that the beam waist would be located at the center of the crystal. The model predicted that using a 125 mm lens, the output would be five times as great. Experimentally, with the same input power, the more pinched waist created by the 125 mm lens did generate more light and was more efficient, but the intensity of the second harmonic was about twice as great. The experimental data and the model are not yet in full agreement and more investigative experiments must be performed to have conclusive data.

As mentioned in Section 5, the efficiency is highest when the greatest integrated intensity volume product of the Gaussian beam is in the crystal. This suggests that if the waist were made smaller by shortening the focal length of the focusing lens and thus shortening the Rayleigh length of the laser beam, the integral intensity may be so concentrated in the center that the amount of second harmonic generation at that point would make up for the beam divergence that occurs more rapidly. The concentrated intensity may be such that it not only compensates for the lack of intensity beyond the Rayleigh length, but would also be able to produce a much more intense second harmonic.

With the preliminary measurements of the second harmonic as the focal length is shortened, it was observed that there is a peak focal length. As the focal length shortens, the output does increase, however, it starts to decrease after the measurements with the 100 mm focal length lens. This means that unlike what was predicted, there is a maximum limit to the extent to how much second harmonic can be produced. It is believed that the con-
centrated beam waist could no longer compensate for the lack of intensity beyond the short Rayleigh length and thus caused the decrease in the total second harmonic output. This explanation for the peak output power also has to be further researched to be conclusive.

An input laser of a higher power could be used in a future experiment to further demonstrate the squared intensity relationship between the input and the output. The frequency doubled light generated by the higher powered input laser could then be more reasonably compared to the second harmonic generated by a pulsed laser.

With the experience gained by doubling infrared light to visible 655 nm light, I could try to find a crystal with a different poling period that could double red light, generating blue light. With that, the blue light can be applied to biotechnology and possibly be used in fluoometry or cytometry.

8 Conclusion

Using the PPLN, a clearly visible second harmonic light of 655 nm was produced with very modest input power. This is significant because the equipment used was neither expensive nor highly specialized and the components were personally gathered and arranged. To generate a second harmonic in a regular LiNbO$_3$ crystal, a high powered, pulsed, and probably rare-earth element doped laser would have to be used. However, with the use of the PPLN and only 5 mW of fundamental light, a second harmonic of $\sim$100 nW was obtained. If the input power were 5 watts, the output power could be a million times larger, or 100 mW. For the first time in the author’s knowledge, the Gaussian beam parameters of the primary light have been adjusted to optimize the second harmonic generation in a periodically poled material. A second harmonic intensity double that found in the confocal geometry was measured, and further improvements should be possible. One can conclude that the use of the PPLN technology for a variety of interesting and beneficial applications is near at hand.
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References


Abstract

Since frequency doubled light can one day offer a cheap, readily available and compact replacement for bulky and expensive Argon lasers, improve biotechnology, and increase the high-density optical storage capacity of compact disks, it has been a subject of considerable interest. A clearly visible beam of red light at 655 nm was successfully generated by frequency doubling a low-power (5 mW) continuous infrared laser light of 1310 nm in a Periodically Poled Lithium Niobate crystal (PPLN).
Doubling the Frequency of Light:
Second Harmonic Generation in Periodically-Poled Lithium Niobate

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