HeXLN: A 2-Dimensional nonlinear photonic crystal

N. G. R. Broderick, G. W. Ross, H. L. Offerhaus, D. J. Richardson and D. C. Hanna
Optoelectronics Research Centre, University of Southampton, Southampton, SO17 1BJ, UK.
Phone: +44 (0)1703 593144, Fax: +44 (0)1703 593142, email: ngr@orc.soton.ac.uk
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We report on the fabrication of what we believe is the first example of a two dimensional nonlinear periodic crystal \[ \text{HeXLN} \], where the refractive index is constant but in which the 2nd order nonlinear susceptibility is spatially periodic. Such a crystal allows for efficient quasi-phase matched 2nd harmonic generation using multiple reciprocal lattice vectors of the crystal lattice. External 2nd harmonic conversion efficiencies > 60% were measured with picosecond pulses. The 2nd harmonic light can be simultaneously phase matched by multiple reciprocal lattice vectors, resulting in the generation of multiple coherent beams. The fabrication technique is extremely versatile and allows for the fabrication of a broad range of 2-D crystals including quasi-crystals.

42.65.K,42.65.-k, 42.70.Qs,42.70.M

The interaction of light with periodic media is an area of intense interest both theoretically and experimentally. A central theme of this work is the idea of a linear photonic crystal \[ \text{(NPC)} \] in which the linear susceptibility is spatially periodic. Photonic crystals can have a complete photonic bandgap over some frequency range and this bandgap can be exploited for a wide variety of processes such as zero threshold lasers, inhibited spontaneous emission, or novel waveguiding schemes such as photonic bandgap fibres \[ \text{(PBGF)} \]. In one dimension photonic crystals, or Bragg gratings, have been well studied for many years. In three dimensions a complete photonic bandgap at long wavelengths has already been demonstrated and work on extending this to the visible region is rapidly progressing \[ \text{(PBGV)} \].

Recently V. Berger proposed extending the idea of photonic crystals to include nonlinear photonic crystals \[ \text{(NPC)} \]. In a nonlinear photonic crystal \[ \text{(NPC)} \] there is a periodic spatial variation of a nonlinear susceptibility tensor while the refractive index is constant. This is in contrast with other work done on nonlinear interactions in photonic crystals \[ \text{(NPC)} \] where the nonlinearity is assumed constant throughout the material and the photonic properties derive from the variation of the linear susceptibility. The simplest type of NPCs are the 1-D quasi-phase-matched materials, first proposed by Armstrong et al. \[ \text{[6]} \] in which the second order susceptibility undergoes a periodic change of sign. This type of 1-D structure has attracted much interest since the successful development of periodically-poled lithium niobate based devices. Generalisation to two and three dimensions in analogy with linear photonic crystals, was recently proposed by Berger and here we report its experimental realisation as a 2-D periodic structure with hexagonal symmetry in lithium niobate \[ \text{(HeXLN)} \].

First we briefly summarise the well known 1-D quasi-phase matching \( \text{(QPM)} \) concept before treating the 2-D case. To this end consider the case of 2nd harmonic generation in a \( \chi^{(2)} \) material where light at a frequency \( \omega \) is converted to a signal at \( 2\omega \). In general the refractive index at \( \omega \) and \( 2\omega \) are different and hence after a length \( L_c \) (the coherence length) the fundamental and the generated 2nd harmonic will be \( \pi \) out of phase. Then in the next coherence length all of the 2nd harmonic is back-converted to the fundamental - resulting in poor overall conversion efficiency. The idea of quasi-phase matching is to change the sign of the nonlinearity periodically with a period of \( L_c \), thus periodically reversing the phase of the generated 2nd harmonic. This ensures that the 2nd harmonic continues to add up in phase along the entire length of the crystal, resulting in a large overall conversion efficiency.

An alternative way to understand the physics of quasi-phase matching is through conservation of momentum. 2nd harmonic generation is a three photon process in which two photons with momentum \( h\kappa \) are converted in a photon of momentum \( h(2\kappa) \) and if \( k^{2\omega} = 2k^\omega \) (ideal phase matching) then the momentum is conserved and the interaction is efficient. However in general due to dispersion ideal phase matching is not possible and different techniques must be used to insure conservation of momentum. In the quasi-phase matched case conservation of momentum becomes \( k^{2\omega} = 2k^\omega + G \), where \( G \) is the crystal momentum corresponding to one of the reciprocal lattice vectors \( \text{(RLV)} \) of the macroscopic periodic structure of the NPC. Clearly this technique allows one to phase-match any desired nonlinear interaction, assuming that one can fabricate an appropriate NPC. In 1-D quasi-phase matching can occur in either the co- or counter-propagating direction. For a strictly periodic lattice quasi-phase matching can only occur over limited wavelength ranges since the RLVs are discrete and periodically spaced in momentum space. In order to obtain broader bandwidths one approach is to use aperiodic structures which have densely spaced RLVs. An alternative approach which is taken here is to move to a two dimensional NPC which brings added functionality compared to a 1-D crystal.

Clearly in a 2-D NPC the possibility of non-collinear phase matching exists due to the structure of the reciprocal lattice. Once again we restrict ourselves to the case of 2nd harmonic generation and linearly polarised light such that we can use the scalar wave equation. Then making the usual slowly varying envelope approximation and assuming...
a plane wave fundamental incident upon the crystal, the evolution equation for the 2nd harmonic in the undepleted pump regime can be written as:

\[ \mathbf{k}^{2\omega} \cdot \nabla E^{2\omega} (\mathbf{r}) = -2i \frac{\omega^2}{c^2} \chi^{(2)}(\mathbf{r}) (E^{\omega})^2 e^{i(k^{2\omega} - 2k^\omega) \cdot \mathbf{r}}. \] (1)

Since \( \chi^{(2)} \) is periodic we can write it as a Fourier series using the RLVs \( \mathbf{G}_{n,m} \)

\[ \chi^{(2)}(\mathbf{r}) = \sum_{n,m} \kappa_{n,m} e^{i \mathbf{G}_{n,m} \cdot \mathbf{r}}, \quad n, m \in \mathbb{Z}. \] (2)

The phase matching condition,

\[ \mathbf{k}^{2\omega} - 2\mathbf{k}^\omega - \mathbf{G}_{n,m} = 0, \] (3)

arises from requiring that the exponent in Eq. (1) be set equal to zero ensuring growth of the 2nd harmonic along the entire length of the crystal. Eq. (1) is a statement of conservation of momentum as discussed earlier. For each RLV \( \mathbf{G}_{n,m} \) and a prescribed \( \mathbf{k}^\omega \) there is at most a unique angle of propagation for the 2nd harmonic such that Eq. (3) is satisfied. The coupling strength of a phase matching process using \( \mathbf{G}_{n,m} \) is proportional to \( \kappa_{n,m} \). If a particular Fourier coefficient is zero then no 2nd harmonic generation will be observed in the corresponding direction.

In order to demonstrate the idea of a 2-D NPC we poled a wafer of lithium niobate with a hexagonal pattern. Fig. 1 shows an expanded view of the resulting structure, which was revealed by lightly etching the sample in acid. Each hexagon is a region of domain inverted material - the total inverted area comprises \( \sim 30\% \) of the overall sample area. The fabrication procedure was as follows. A thin layer of photoresist was first deposited onto the \(-z\) face of a 0.3mm thick, \(z\)-cut wafer, of LiNbO\(_3\), and then photolithographically patterned with the hexagonal array. The \(x-y\) orientation of the hexagonal structure was carefully aligned to coincide with the crystal’s natural preferred domain wall orientation: LiNbO\(_3\) itself has triagonal atomic symmetry (crystal class 3m) and shows a tendency for domain walls to form parallel to the \(y\)-axis and at \(\pm 60^\circ\) as seen in Fig. 1. Poling was accomplished by applying an electric field via liquid electrodes on the +/-z faces at room temperature. Our HeXLN crystal has a period of 18.05 \(\mu\)m: suitable for non-collinear frequency doubling of 1536nm at 150\(^\circ\)C (an elevated temperature was chosen to eliminate photorefractive effects). The hexagonal pattern was found to be uniform across the sample dimensions of 14 \(\times\) 7mm (\(x-y\)) and was faithfully reproduced on the +z face. Lastly we polished the \(\pm x\)-faces of the HeXLN crystal allowing a propagation length of 14mm through the crystal in the \(\Gamma K\) direction (see Fig. 1).

In Fig. 2 we show the reciprocal lattice (RL) for our HeXLN crystal. In contrast with the 1-D case there are RLVs at numerous angles, each of which allows phase matching in a different direction (given by Eq. 3). Note that for a real space lattice period of \(d\) the RL has a period of \(4\pi/\sqrt{3}\) as compared with \(2\pi/d\) for a 1-D crystal allowing us to compensate for a greater phase mismatch in a 2-D geometry than in a 1-D geometry with the same spatial period. From Eq. (3) and using simple trigonometry it is possible to show that

\[ \frac{\lambda^{2\omega}}{n^{2\omega}} = \frac{2\pi}{|\mathbf{G}|} \sqrt{\left(1 - \frac{n^{2\omega}}{n^{2\omega} \sin^2 \theta}\right)^2 + 4 \frac{n^{2\omega}}{n^{2\omega} \sin^2 \theta}}, \] (4)

where \(\lambda^{2\omega}\) is the vacuum wavelength of the second harmonic and \(2\theta\) is the walk off angle between the fundamental and 2nd harmonic wavevectors.

To investigate the properties of the HeXLN crystal we proceeded as follows. The HeXLN crystal was placed in an oven and mounted on a rotation stage which could be rotated by \(\pm 15^\circ\) around the \(z\)-axis while still allowing light to enter through the \(+z\) face of the crystal. The fundamental consisted of 4ps, 300kW pulses obtained from a high power all-fibre chirped pulse amplification system (CPA) operating at a pulse repetition rate of 20kHz. The output from the CPA system was focussed into the HeXLN crystal using a 10cm focal length lens giving a focal spot diameter of 150\(\mu\)m and a corresponding peak intensity of \(\sim 1.8\text{GW/cm}^2\). The initial experiments were done at zero angle of incidence corresponding to propagation in the \(\Gamma K\) direction. At low input intensities \((\sim 0.2\text{GW/cm}^2)\) the output was as shown in Fig. 2(b) and consisted of multiple output beams of different colours emerging from the crystal at different angles. In particular two 2nd harmonic beams emerged from the crystal at symmetrical angles of \(\pm (1.1 \pm 0.1)^9\) from the remaining undeflected fundamental. At slightly wider angles were two green beams (third harmonic of the pump) and at even wider angles were two blue beams (the fourth harmonic, not shown here). There was also a third green beam copropagating with the fundamental. The output was symmetrical since the input direction corresponded to a symmetry axis of the NPC. As the input power increased the 2nd harmonic spots remained in the same positions while the green light appeared to be emitted over an almost continuous range of angles rather than the discrete angles.
observed at low powers. The two 2nd harmonic beams can be understood by referring to the reciprocal lattice of our structure (Fig. 2). From Fig. 3 it can be seen that for propagation in the ΓK direction the closest RLVs are in the ΓM directions and it is these RLVs that account for the 2nd harmonic light.

After filtering out the other wavelengths the 2nd harmonic (from both beams) was directed onto a power meter and the efficiency and temperature tuning characteristics for zero input angle were measured. These results are shown in Fig. 3 and Fig. 4. Note that the maximum external conversion efficiency is greater than 60% and this is constant over a wide range of input powers. Taking into account the Fresnel reflections from the front and rear faces of the crystal this implies a maximum internal conversion efficiency of 82% – 41% in each beam. As the 2nd harmonic power increases the amount of back conversion increases which we believe is the main reason for the observed limiting of the conversion efficiency at high powers.

Evidence of the strong back conversion can be seen in Fig. 4 which shows the spectrum of the remaining fundamental for both vertically (dashed) i.e. in the z-direction and horizontally (solid line) polarised input light. As the phase matching only works for vertically polarised light the horizontally polarised spectrum is identical to that of the input beam and when compared with the other trace (dashed line) shows the effect of pump depletion and back-conversion. Note that for vertically polarised light the amount of back-converted light is significant compared to the residual beam and when compared with the other trace (dashed line) shows the effect of pump depletion and back-conversion.

In the 1-D case, for an undepleted pump, the temperature tuning curve of a 14mm long length of periodically poled material is expected to have a \( \text{sinc}^2(T) \) shape and to be quite narrow – 4.7°C for a 1-D PPLN crystal with the same length and period as the HeXLN crystal used here. However, as can be seen from Fig. 5 the temperature tuning curve (obtained in a similar manner to the power characteristic) is much broader with a FWHM of 25°C, and it exhibits considerable structure. The input power was 300kW. We believe that the increased temperature bandwidth may be due to the multiple reciprocal lattice vectors that are available for quasi-phase matching with each RLV producing a beam in a slightly different direction. Thus the angle of emission of the 2nd harmonic should vary slightly with temperature if this is the case. Due to the limitations of the oven we were not able to raise the temperature above 205°C and hence could not completely measure the high temperature tail of the temperature tuning curve. Note that temperature tuning is equivalent to wavelength tuning of the pump pulse and hence it should be possible to obtain efficient phase-matching over a wide wavelength range at a fixed temperature.

After the properties of the HeXLN crystal at normal incidence we next measured the angular dependance of the 2nd harmonic beams. As the crystal was rotated phase-matching via different RLVs could be observed. For a particular input angle (which determined the angle between the fundamental and the RLVs) quasi-phase matched 2nd harmonic generation occurred, via a RLV, and produced a 2nd harmonic beam in a direction given by Eq. (4). These results are shown in Fig. 6 where the solid circles indicate the measured angles of emission for 2nd harmonic light while the open squares are the predicted values. In the figure zero degrees corresponds to propagation in the ΓK direction. Also indicated on the figure are the RLVs used for phase-matching, where \([n,m] \) refers to the RLV \( G_{n,m} \). Note that there is good overall agreement between the theoretical and experimental results even for higher order Fourier coefficients which indicates the high quality of the crystal. The inversion symmetry of Fig. 6 results from the hexagonal symmetry of the crystal. To further highlight this symmetry we have labeled the negative output angles with the corresponding positive RLVs. The only obvious discrepancy comes from the \([1,1] \) RLVs where two closely separated spots are observed rather than a single one. This may be due to a small amount of linear diffraction from the periodic array. At the domain boundaries of the HeXLN crystal there are likely to be small stress-induced refractive index changes giving a periodic variation in the refractive index. If this indeed proves to be the case then it should be possible to eliminate this by annealing the crystal at high temperatures.

For applications where collinear propagation of the fundamental and 2nd harmonic is desirable propagation along the \( \Gamma M \) axis of the HeXLN crystal could be used (since the smallest RLV is in that direction). For the parameters of our crystal collinear 2nd harmonic generation of 1.44μm in the \( \Gamma M \) direction is expected.

Visually the output of the HeXLN crystal is quite striking with different colours (red, green and blue) being emitted in different directions (see Fig. 7). For a range of input angles and low powers distinct green and red spots can been seen each emitted in a different direction, often with the green light emitted at a wider angle than the 2nd harmonic. The presence of the green light implies sum frequency generation between the fundamental and the 2nd harmonic. For this to occur efficiently it must also be quasi-phase-matched using a RLV of the lattice. In certain regimes (of angle and temperature) simultaneous quasi-phase-matching of both 2nd harmonic generation and sum frequency mixing occurs with as much as 20% of the 2nd harmonic, converted to the green (in multiple beams). As mentioned earlier at higher powers the green light appears to be emitted over a continuous range of angles. We believe that this might be due to an effect similar to that observed in fibres where phase-matching becomes less critical at high intensities.
It should be noted that although lithium niobate preferentially forms domains walls along the $y$ axis and at $\pm 60^\circ$ we are not limited to hexagonal lattices. In fact essentially any two dimensional lattice can be fabricated, however the patterned region of the unit cell will always consist of either a hexagon or a triangle. The shape of the poled region will determine the strength of each of the Fourier coefficients for the RLVs while the lattice structure will determine their position. One can envisage creating more complicated structures such as a 2-D quasi-crystal in which a small poled hexagon is situated at every vertex. Such a 2-D quasi-crystal could give improved performance for simultaneously phase matching multiple nonlinear processes, as demonstrated recently with a 1-D poled quasi-crystal \[12\]. Alternatively a HeXLN crystal could be used as an efficient monolithic optical parametric oscillator \[1\]. Lastly we note that NPCs are a specific example of more general nonlinear holographs which would convert a beam profile at one wavelength to an arbitrary profile at a second profile \[13\]. For example Imeshevx et al. converted a gaussian profile beam at the fundamental to a square top 2nd harmonic using transversely patterned periodically poled lithium niobate \[14\].

In conclusion we have fabricated what we believe to be the first example of a two dimensional nonlinear photonic crystal in Lithium Niobate. Due to the periodic structure of the crystal, quasi-phase matching is obtained for multiple directions of propagation with internal conversion efficiencies of $> 80\%$. Such HeXLN crystals could find many applications in optics where simultaneous conversion of multiple wavelengths is required.

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\textbf{FIG. 1} . Picture of the HeXLN crystal and the first Brillouin zone. The period of the crystal is 18.05 $\mu$m and is uniform over the whole sample. In our experiments propagation was in the $\Gamma K$ direction.

\textbf{FIG. 2} . Reciprocal Lattice for the hexagonal lattice shown in Fig. 1. The general reciprocal lattice vector $\mathbf{G}_{n,m} = n\mathbf{e}_1 + m\mathbf{e}_2$ where $\mathbf{e}_1, \mathbf{e}_2$ are the basis vectors for the reciprocal lattice. Also indicated is the first Brillouin zone showing the main symmetry directions. In addition two examples of non-collinear QPM are shown using the $[1,0]$ and the $[1,1]$ RLVs. On the right is a picture of the low power output of the crystal. Note that there are two 2nd harmonic spots and three 3rd harmonic spots.
FIG. 3. 2nd harmonic efficiency of the HeXLN crystal against input peak power. Note that the maximum efficiency is > 60% and is limited principally by parametric back conversion.

FIG. 4. Output spectra at 1533nm for both horizontally (solid line) and vertically (dashed line) polarised light. Note the large amount of pump depletion which can clearly be seen along with the back-conversion. The incident peak power was 300kW.

FIG. 5. Temperature tuning of the HeXLN crystal taken at an incident peak power of 300kW. The temperature tuning curve is much broader than a comparable 1-D PPLN crystal and posses multiple features has to the large number of reciprocal lattice vectors available.

FIG. 6. Graph of the experimental (circles) and theoretical (squares) output angles for the 2nd harmonic as an function of the external input angle, where $0^\circ$ indicates propagation in the $\Gamma K$ direction. The maximum internal angle between the fundamental and 2nd harmonic was $\sim 8^\circ$ (the refractive index of lithium niobate is $\sim 2.2$).

FIG. 7. Output of the HeXLN crystal at high powers and a variety of input angles.
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