

HeXLN: A 2-Dimensional nonlinear photonic crystal

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Abstract

We report on the fabrication of what we believe is the first example of a two dimensional nonlinear periodic crystal [1], 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 would allow 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

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 $\hbar k^\omega$ are converted in a photon of momentum $\hbar k^{2\omega}$ and if $k^{2\omega} = 2k^\omega$ (ideal phase matching) then the momentum is conserved and the interaction is allowed. 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 (RLV) [7] 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 [1]:

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

the HeXLN crystal allowing a propagation length of 14mm through the crystal in the ΓK 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 $4\pi/(\sqrt{3}d)$ as compared with $2\pi/d$ for a 1-D crystal [9] 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 [1]

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

where $\lambda^{2\omega}$ is the vacuum wavelength of the second harmonic and 2θ 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 $+x$ face of the crystal. The fundamental consisted of 4ps, 300kW pulses obtained from a high power all-fibre chirped pulse amplification system (CPA) [10] 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 $150\mu\text{m}$ and a corresponding peak intensity of $\sim 1.8\text{GW}/\text{cm}^2$. The initial experiments were done at zero angle of incidence corresponding to propagation in the ΓK direction. At low input intensities ($\sim 0.2\text{GW}/\text{cm}^2$) 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 $\pm(1.1 \pm 0.1)^\circ$ 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

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 $\sim 25^\circ\text{C}$, 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 dependence 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 single 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 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 $\mathbf{G}_{n,m}$. 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

determine their position. One can envisage creating more complicated structures such as a 2-D quasi-crystals 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 [13]. 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 [14]. For example Imeshev *et al.* converted a gaussian profile beam at the fundamental to a square top 2nd harmonic using transversely patterned periodically poled lithium niobate [15].

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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