

Characterisation of Potassium Titanyl Phosphate and Potassium Niobate Crystals in an Infrared OPO

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Executive Summary

The efficiency of potassium titanyl phosphate (KTP) and potassium niobate crystals in a two-mirror optical parametric oscillators (OPOs) has been measured in order to assess their suitability for use in some atmospheric monitoring applications.

This work forms a small part of the output from the Brite-Euram NOCTILS project.

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1. INTRODUCTION

The performance of various non-linear optical crystals was assessed at NPL as part of a project funded under the European Union's BRITE-EURAM Programme to develop new optical materials suitable for the generation of pulsed, tunable infrared radiation (NOCTILS). The specific application area that was targeted for these new materials was as an all-solid-state source for the next generation of infrared differential absorption lidar (DIAL) systems, an area in which NPL has extensive experience¹.

The optical source should produce narrow bandwidth radiation continuously tunable at least across the range 3.1 μm to 3.6 μm in order to be useful for infrared DIAL applications. This is the region of the fundamental carbon-hydrogen stretch absorption which is the key spectroscopic signature for many industrially-important species. The optical sources discussed in this report are all designed to operate in the near-infrared region. The target wavelengths in the mid-infrared would be obtained by mixing the near-infrared output with a 1 μm pump in an optical parametric amplifier (OPA). This mixing mechanism determines the wavelength specification in the near-infrared, which is a narrow bandwidth source tunable between 1.5 μm and 1.6 μm .

An optical parametric oscillator (OPO) was developed at NPL specifically for the purpose of testing potassium niobate (KNbO_3) and potassium titanyl phosphate (KTP) crystals supplied by two members of the consortium, F.E.E. and CRISTAL LASER. The suitability of another non-linear material, lithium niobate, had already been investigated in previous research conducted at NPL^{2,3}. The OPO developed for this project used two plane mirrors (input coupler : HR @ 1.55 μm , HT @ 1 μm ; output coupler 70%T @ 1.55 μm , HT @ 1 μm) and was operated with a variable spacing between them. This enabled the efficiency of the crystals to be measured as a function of cavity length, which then facilitated the prediction of their performance in specific OPO systems.

The crystal performance parameters that were investigated were the oscillation threshold and efficiency, the ability to predict and control the output wavelength of the OPO, and the potential for the rapid fine-scale wavelength tuning required for DIAL operation.

2. CHARACTERISATION OF POTASSIUM NIOBATE CRYSTALS

Eleven KNbO_3 samples were received by NPL for testing during the project and a summary of the data obtained is given in Table 2.1 below.

Table 2.1 : Potassium niobate crystals tested at NPL

KNbO ₃ Reference	Signal (mJ)	Pump (mJ)	Cavity length (mm)
A109-23/2	1	80	20
C109-13/1	10	66	20
C109-13/2	9.5	66	20
C109-13/3	12	66	20
A109-24	8	66	20
D102-12	10	66	20
D96-6 (Damaged)	-	66	20
A113-3	8	100	20
A126-16C	6	100	30
	9	100	10
A126-16D	1	66	30
162-7/1	2	58	20

The phase-matching curve for potassium niobate in an OPO pumped at 1064 nm is shown in Figure 1. These data are derived from the Sellmeier coefficients derived by Zysset⁴.

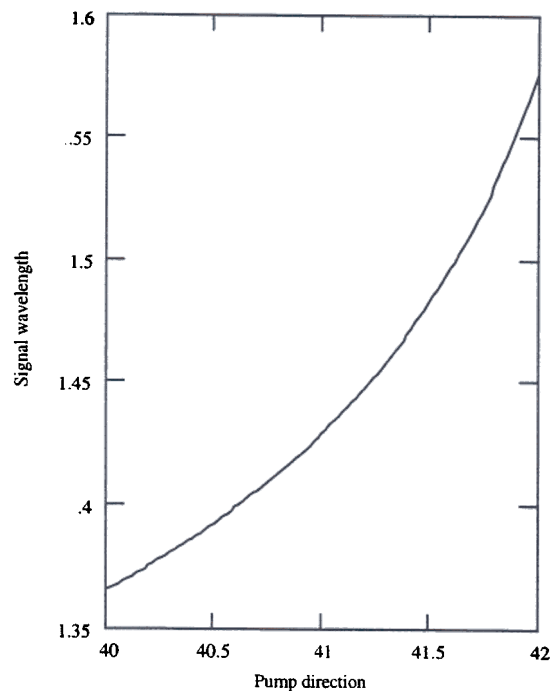


Figure 1 : Phase-matching curve for KNbO_3 OPO with 1064 nm pump

Figure 2 shows an example of the relationship between pump energy and signal energy for one of the potassium niobate crystals (C109-13/1). A linear fit has been applied to the data to determine the threshold and slope efficiencies, indicating in this case a threshold of 18 mJ, and a slope efficiency of 27%. The same crystal was tested in a 40 mm cavity which gave a factor of two reduction in output energy. This graph is typical of the measurements made at NPL on potassium niobate crystals.

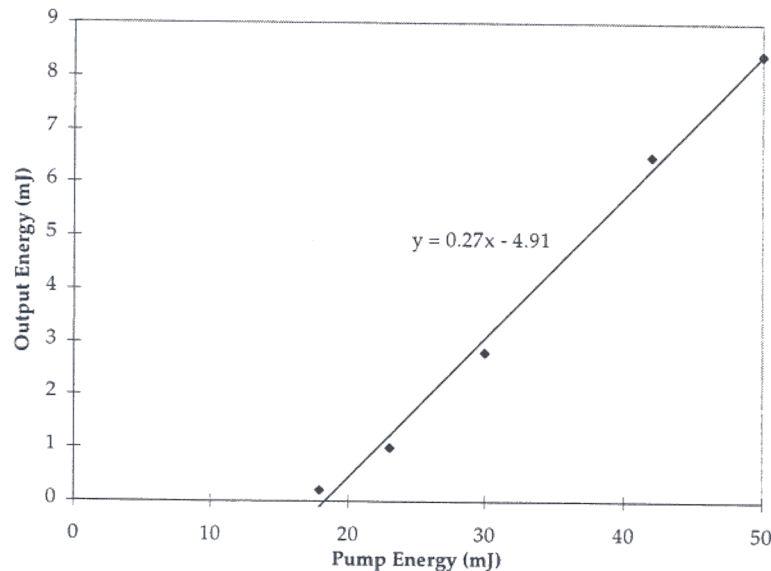


Figure 2 : Slope efficiency and threshold of a potassium niobate crystal

The potential of potassium niobate for use as an optical parametric amplifier was also investigated. Crystals A126-16C and D were fitted after a KTP OPO producing a signal energy of 4-5 mJ at 1.5 μm , and pumped with 170 mJ (5 mm diameter) of 1064 nm radiation. No detectable amplification was observed. The standard OPA used by NPL for infrared DIAL applications (which uses a 50 mm long lithium niobate crystal)⁵ produced 12 mJ of amplified signal in the same configuration.

Another feature of the potassium niobate crystals used in these experiments was their susceptibility to optically-induced damage. Almost all of the crystals tested showed visible surface and/or bulk damage after long term (4+ hours) operation at the pumping levels required for stable oscillation.

3. CHARACTERISATION OF KTP CRYSTALS

A series of KTP crystals have also been characterized. KTP is a biaxial crystal and can be usefully phase-matched in three different planes for the applications considered by NOCTILS. Figure 3 shows the theoretical phase matching angle for 1064 nm pumped and 532 nm pumped OPOs with the zx, xy and yz planes displayed continuously.

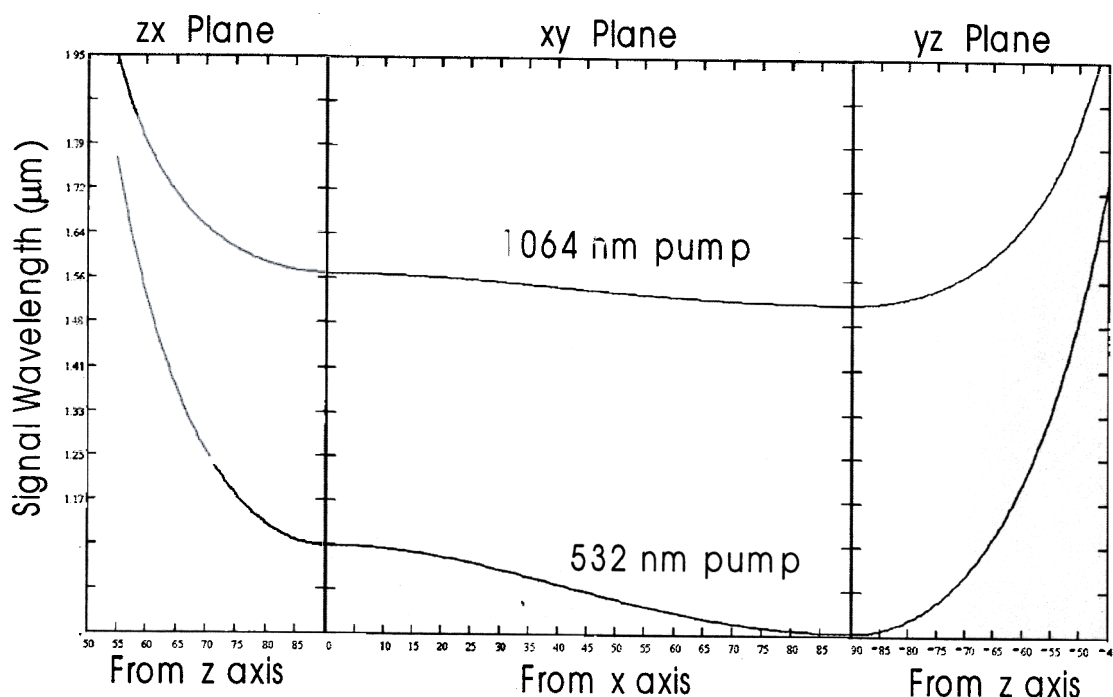


Figure 3 : Phase matching curves for KTP OPO
with 1064 nm and 532 nm pump wavelengths

3.1. CRYSTAL PERFORMANCE

Table 2.2 shows the results of the performance tests carried out on nine KTP crystals with a 1064 nm pumped 2-mirror OPO in collinear configuration. The theoretical collinear signal wavelengths given in this Table have been calculated using the Sellmeier coefficients given by Bierlein⁶. The signal energy slope efficiency and the threshold pump energy are for a 35 mm long plane-plane cavity with a 2.5 mm diameter pump beam, except for the starred (*) cases for which the cavity length was 27 mm.

Table 2.2 : Summary of results of KTP crystal tests at NPL

Crystal Description	Crystal Reference	Crystal size (mm)	Collinear Wavelength (nm)		Slope Efficiency	Threshold (mJ)
			Theoretical	Measured		
X-cut $\phi=0$	62 713 027	5x5x21	1571	1572	30%	11
XY-cut $\phi=18.5$	63 309 003	5x5x20	1565	1567	29%	23
XY-cut $\phi=23.5^*$	82 302 073	5x5x20	1562	1564	42%	22
XY-cut $\phi=24.7$	71 109 055	6x6x10	1561	1563	28%	50
XY-cut $\phi=32.0^*$	84 813 076	8x8x21	1556	1558	-	-
XY-cut $\phi=34.7$	80 613 181	3x3x20	1553	1556	14%	51
XY-cut $\phi=49.9$	82 909 011	5x8x20	1540	1543	39%	32
XY-cut $\phi=50.8$	71 109 040	5x5x10	1539	1552	-	65
XZ-cut $\theta=45.5$	70 807 003	6x6x20	1689	1690	24%	25

Figure 4 gives some examples of the data used to derive the threshold and efficiency values for the KTP crystals.

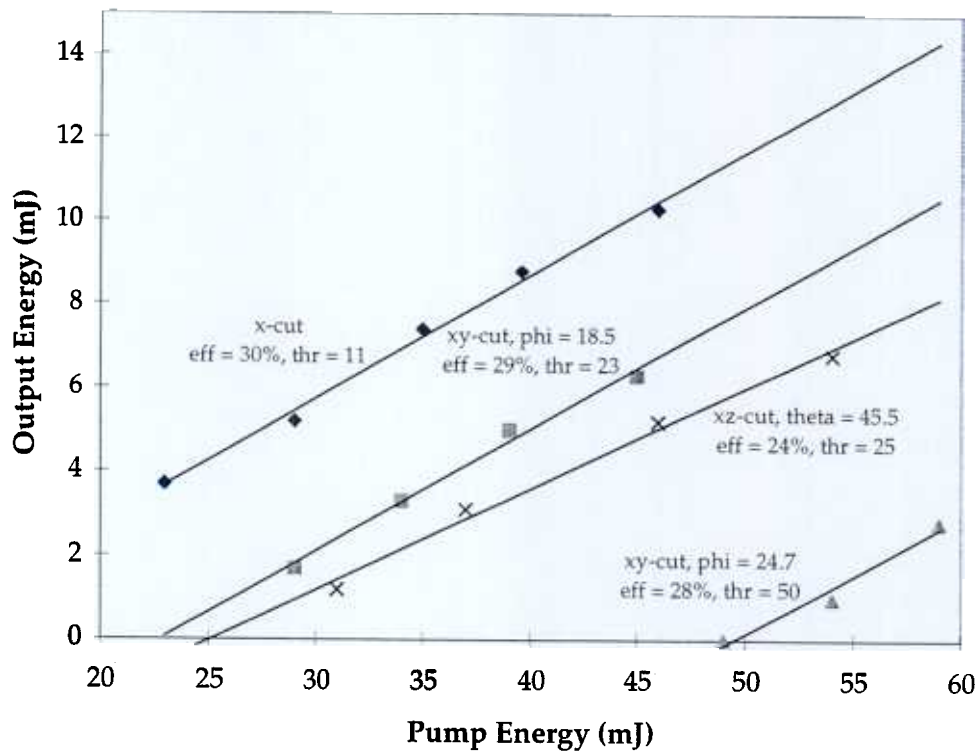


Figure 4 : Slope efficiency and threshold measurements of xy-cut KTP crystals

3.2. WAVELENGTH MEASUREMENTS

The signal wavelengths were measured using the experimental arrangement shown schematically in Figure 5.

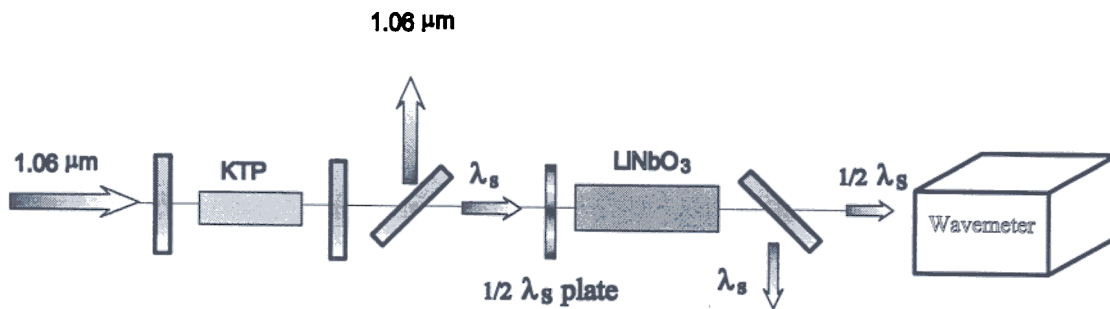


Figure 5 : Schematic layout of KTP oscillation wavelength measurement (collinear)

The output from the OPO was frequency doubled in a lithium niobate crystal so that the wavelength could be measured in a pulsed wavemeter (Burleigh PWA-4500). A clear discrepancy between the measured and theoretical values was observed. Figure 6 shows the relationship between crystal angle and the collinear wavelength, both for the measured values and two sets of theoretical values, one derived from the Bierlein Sellmeier coefficients⁶ and the other from the coefficients derived by Kato⁷.

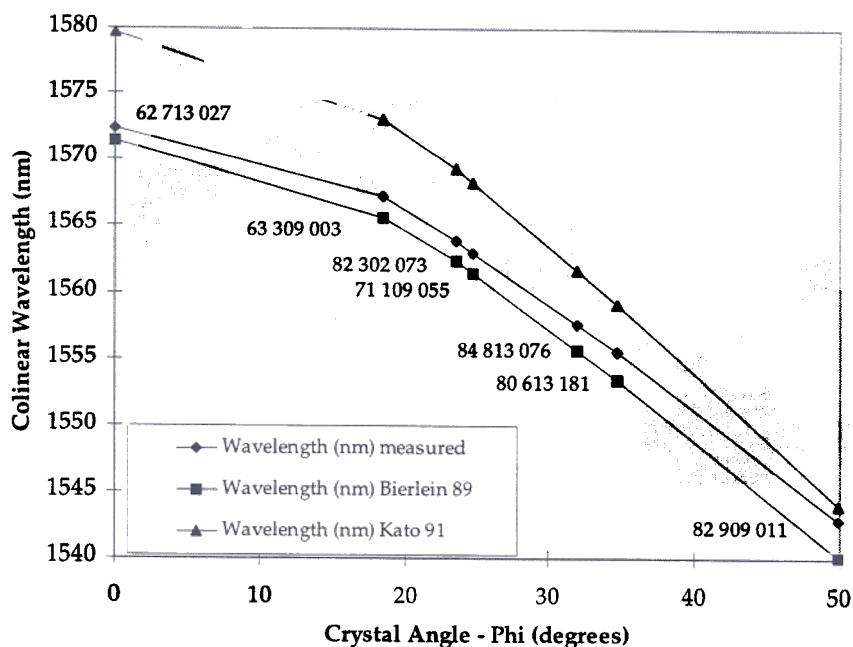


Figure 6 : Oscillation wavelength for xy-cut KTP OPO's

Figure 7 shows that the discrepancy between the measured and theoretical (Bierlein) values shows an approximately linear form with increasing crystal angle in the XY plane. The linear fit shown on this graph enables the required crystal angle (in the XY plane) to be specified for any given signal wavelength.

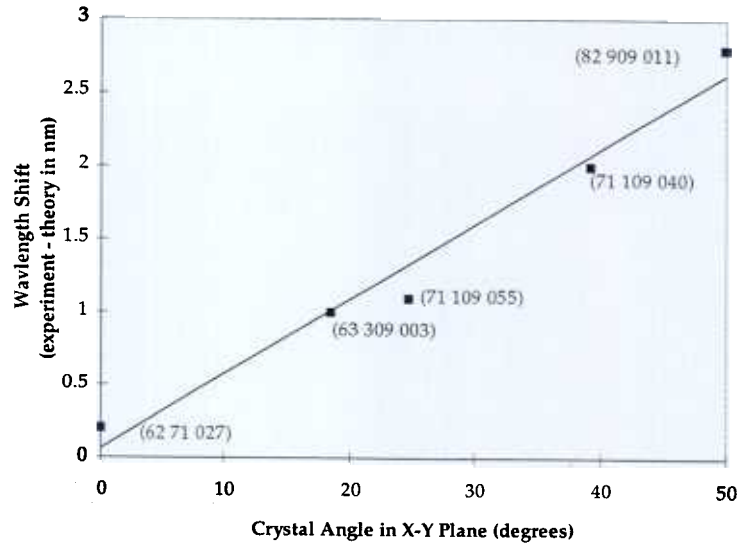


Figure 7 : Experimental and theoretical oscillation wavelengths for xy-cut KTP (in collinear geometry)

3.3. FINE-SCALE WAVELENGTH TUNING

After testing the collinear oscillation wavelengths of crystals cut at different angles, the fine scale tunability of individual crystals was investigated. Two different mechanisms were studied - crystal angle tuning, and non-collinear angle tuning. Crystal angle tuning involves rotating the crystal within the OPO cavity, while leaving the pump and signal beam directions unchanged. The measured tuning rate was 0.57 nm per degree which agrees well with the theoretical value as shown in Figure 8.

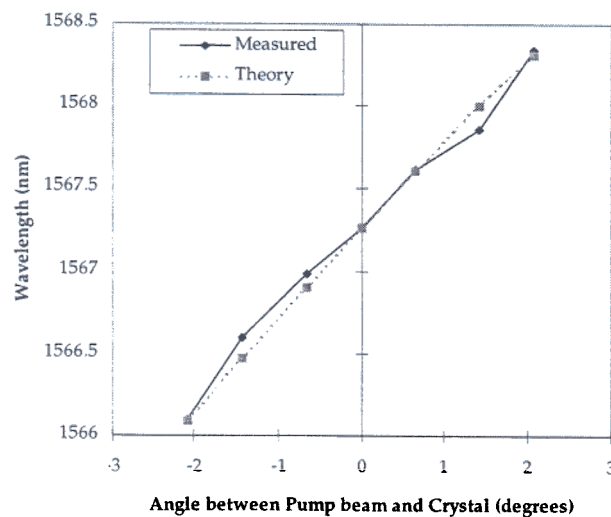


Figure 8 : Variation of wavelength with crystal angle for 18.5 degree xy-cut KTP crystal (#63 309 003)

Non-collinear angle tuning involves changing the angle between the pump and signal beam, and is achieved by changing the angle at which the pump beam enters the OPO cavity. Figure 9 shows the schematic phase match diagram for non-collinear operation. If the non-collinear angle α is changed, the length of the signal and idler vectors k_s and k_i will also have to change to maintain phase matching, and hence change the signal and idler wavelengths.

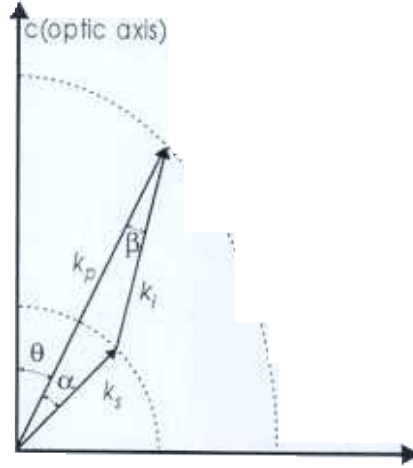


Figure 9 : Orientation of the beams for non-collinear phase matching

Figure 10 shows the agreement between measured and theoretical tuning behaviour, and also highlights two important features of non-collinear angle tuning in KTP. Firstly, that increasing angle produces longer wavelengths independently of the sign of the angle, and secondly that the rate of tuning is significantly higher than the rate for crystal angle tuning.

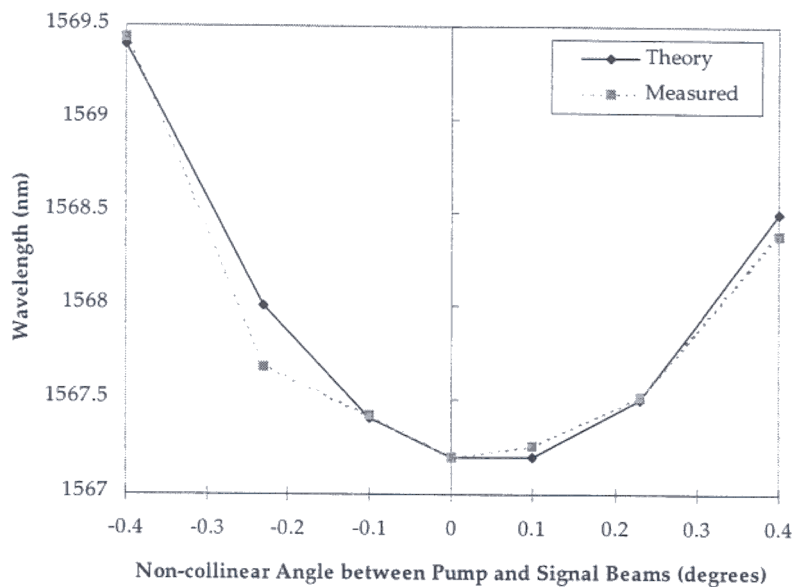


Figure 10 : Variation of wavelength with non-collinear angle for 18.5 degree xy-cut KTP (crystal # 63 309 003)

4. CALCULATION OF PHASE-MATCHING ACCEPTANCE ANGLES

The key parameters that differentiate the performance of different nonlinear crystals in particular applications include the three possible acceptance angles:

- The crystal acceptance angle ($\Delta\vartheta_c$) - is the angle through which the crystal can be rotated with only a specified reduction in efficiency with the pump and signal fixed in space.

The signal acceptance angle ($\Delta\vartheta_s$) - is the angle through which the crystal can be rotated with only a specified reduction in efficiency with only the pump fixed in space.

- The pump acceptance angle ($\Delta\vartheta_p$) - is the angle through which the crystal can be rotated with only a specified reduction in efficiency with only the signal fixed in space.

We calculate the three possible acceptance angles numerically by solving the equation for the mismatch in the generated (idler) wave:

$$(k_i + \Delta k)^2 = k_p(\vartheta)^2 + k_s^2 - 2 * k_p * k_s * \cos(\alpha)$$

The acceptance angle is given by the solution of this equation for a different combination of angles for a stated value of $\Delta k = \pi/l$. For example:

the crystal acceptance angle -

$$(k_i + \pi/l)^2 = k_p(\vartheta + \Delta\vartheta_c)^2 + k_s^2 - 2 * k_p * k_s * \cos(\alpha)$$

the signal acceptance angle -

$$(k_i + \pi/l)^2 = k_p(\vartheta)^2 + k_s^2 - 2 * k_p * k_s * \cos(\alpha + \Delta\vartheta_s)$$

the pump acceptance angle -

$$(k_i + \pi/l)^2 = k_p(\vartheta + \Delta\vartheta_p)^2 + k_s^2 - 2 * k_p * k_s * \cos(\alpha - \Delta\vartheta_p)$$

Each of these three angles has been calculated by NPL for KNbO_3 , LiNbO_3 , and KTP (non-critically phase matched). These calculations are particularly helpful in quantifying the advantages of KNbO_3 over LiNbO_3 . The crystal acceptance angle of KNbO_3 is 0.8 mrad for a 10 mm crystal which is smaller than for LiNbO_3 (1.4 mrad). Tangential phase matching⁸ is possible at angles between 1.2 and 1.6 degrees (depending on the signal wavelength). This angle is twice as large as for LiNbO_3 , which reflects the larger walk-off angle in KNbO_3 .

Figures 11 and 12 show the acceptance angles for 10 mm samples of KNbO_3 and LiNbO_3 . The curves for idler wavelengths of 1.5 μm are also shown. The large values of $\Delta\vartheta_p$ correspond to the tangential phase matching condition.

Issues related to acceptance angles were the topic of an ongoing discussion inside the NOCTILS consortium. Using the code developed by NPL the data summarized below are obtained in the case of a LiNbO_3 OPO pumped at 1064 nm with a signal at 1500 nm and an idler at 3660 nm. Also included are the results obtained with the same code for KNbO_3 and KTP (with a “fictive” idler). The results are summarized in Table 2.3. These results are very similar to those generated by a separate code developed by A. Smith (Sandia National Laboratory, USA).

Table 2.3 : Characterisation of different crystals in OPO configuration

	LiNbO_3	KNbO_3	KTP
Temperature	300 K	300 K	300 K
Pump (nm)	1064(e)	1064(e)	1064(o)
Idler + Signal (nm)	3661(o) + 1500(o)	3661(o) + 1500(o)	3661(e) + 1500(e)
Orientation (deg)	Theta = 47.1	Theta = 41.5 Phi = 0	Theta = 44.3 Phi = 0
d_{eff} (pm/V)	-4.92	-9.65	-2.52
Walkoff (mrad)	34	67	51
Accept, angle (mrad-cm)	1.4	0.8	1.7
Accept, bandwidth (cm^{-1} -cm)	23	23	180
Temp, accept, (K-cm)	4.7	16.7	54

4.1 POLYCHROMATIC PHASE MATCHING

One particular phase matching condition that is of potential interest in this type of application is polychromatic phase matching⁹. The requirements to meet this condition are discussed in Appendix 1. The main feature of polychromatic phase matching is that when this condition is met the output bandwidth of the OPO is very wide, potentially covering the entire wavelength range of interest. Line narrowing techniques, such as injection seeding, could then be used to select the exact wavelength required for a particular measurement without having to make any adjustments to the OPO. However, calculations of the polychromatic phase matching conditions for KPT and potassium niobate showed that the appropriate conditions could not be met in the wavelength range of interest.

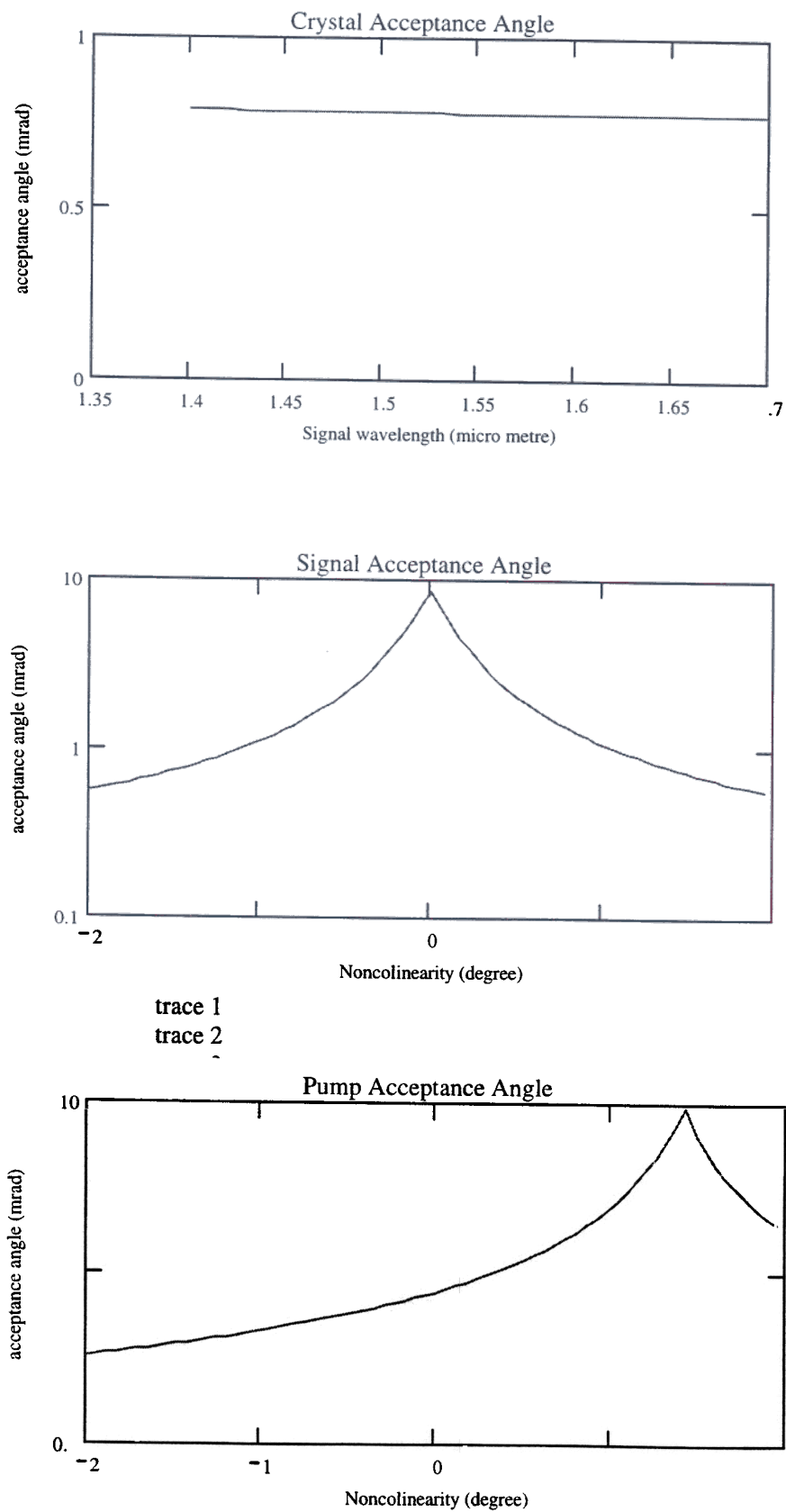


Figure 11 Acceptance Angles for Potassium Niobate (10 mm sample)

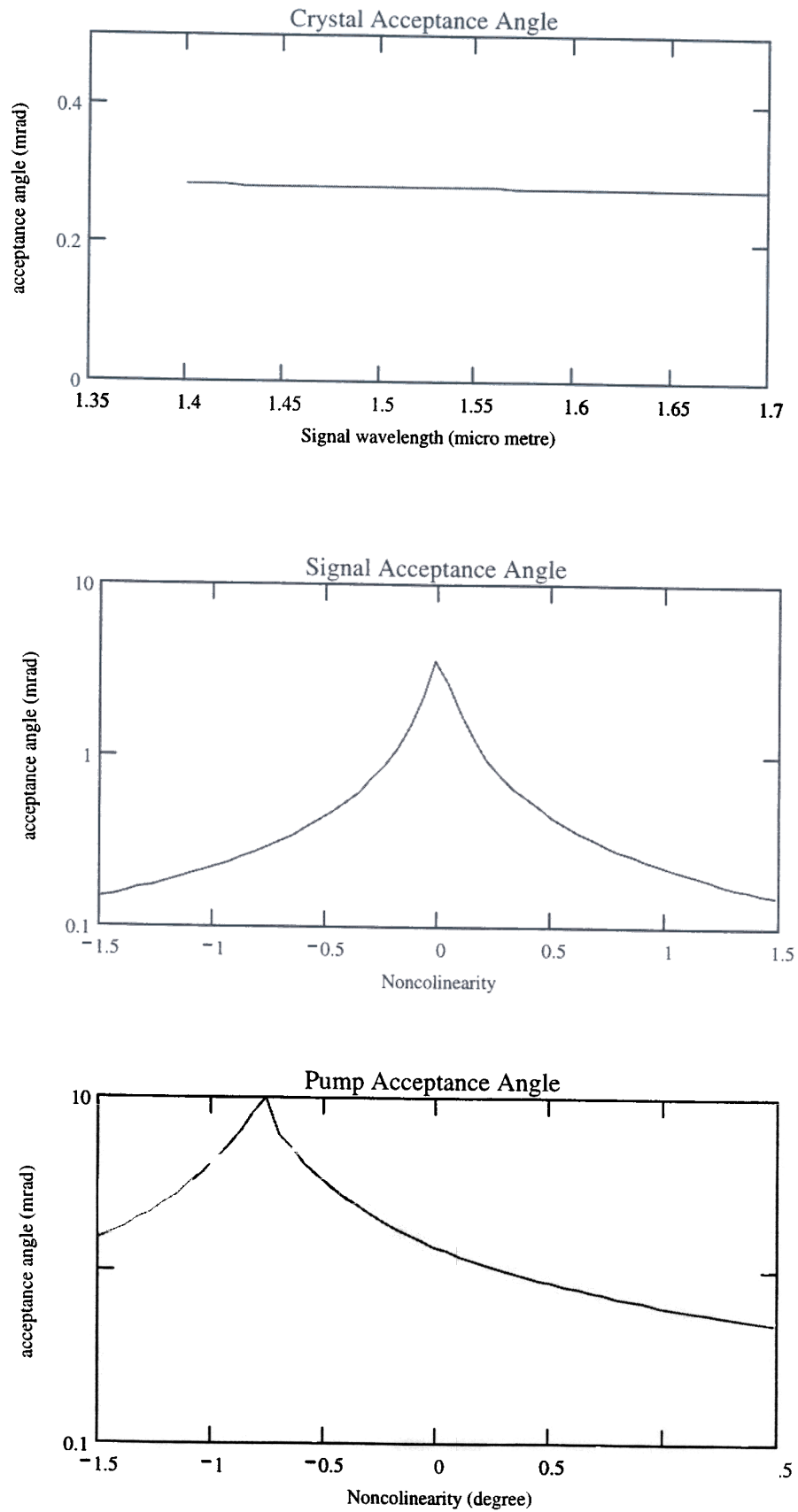


Figure 12 Acceptance Angles for Lithium Niobate (50 mm sample)

5. INVESTIGATION OF 532 NM PUMPED OPO

All of the work described in the previous sections deals with a pump wavelength of 1064 nm. An alternative pumping scheme was also investigated, in which a two-mirror OPO was pumped at 532 nm. The OPO cavity was set up to be resonant at the signal wavelength (around 810 nm) and fully transmitting at the idler wavelength (around 1520 nm). The idler output could then be used as the input to a 1064 nm pumped OPA, which would then give the required infrared output at 3 μm .

As can be seen in Figure 3, the KTP crystal cut that would be used to obtain an output in the required signal range (XZ cut KTP) has a high angular tuning rate. This gives the major benefit that a single crystal would cover the entire range of target wavelengths.

Initial tests were carried out with a short (10 mm long) uncoated KTP crystal. Unseeded OPO operation was demonstrated in a 13 mm long two-mirror cavity with a 33% slope efficiency and a pump threshold of 13 mJ. The crystal angle tuning behaviour of the 532 nm pumped OPO was measured, and this agreed well with the theoretical prediction, as shown in Figure 13. Injection-seeded operation has also been demonstrated, and the wide tuning range of 1507 to 1548 nm confirmed with narrow bandwidth operation.

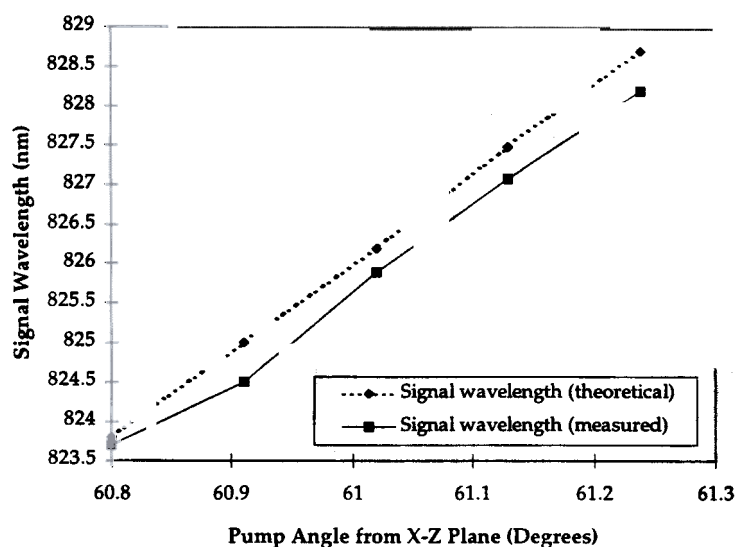


Figure 13 : Tuning curve of 532 nm pumped OPO

Following the successful initial testing of the 532 nm pumped OPO, a crystal was supplied by CRISTAL LASER that met the specific requirements for infrared DIAL operation. This 20.6 mm long crystal was cut at $\theta = 59.3^\circ$ and $\phi = 0^\circ$ to give tuning across the required wavelength range. In addition it was AR coated at pump, signal and idler wavelengths to prevent losses and residual oscillation. The crystal was tested in a 32 mm long two-mirror cavity and was shown to have a combined signal+idler slope efficiency of 50% and a pump threshold of 5.5mJ (with a 2.5 mm diameter pump). The collinear signal wavelength was 809.68 nm. The new crystal was successfully injection seeded at various (idler) wavelengths between 1520 and 1547 nm, which cover the range required for useful infrared DIAL measurements.

6. CONCLUSIONS

- Potassium niobate had been used successfully in OPO's with similar performance to that demonstrated previously for lithium niobate. Most samples of potassium niobate exhibited some bulk damage after a few hours of operation, which limited the scope for detailed investigation of the potential for its use.
- The performance of various KTP crystals was assessed using a simple 2-mirror OPO cavity pumped at 1064 nm. Output slope efficiencies varied between 14% and 42%, with oscillation thresholds varying between 11 mJ and 65 mJ.
- The collinear output wavelength of the OPO was measured with a series of XY-cut KTP crystals. A significant discrepancy was observed between the measured values and the two sets of available theoretical data. The relationship between the discrepancy and the crystal angle was determined, and this enables the collinear wavelength for any given XY-cut KTP crystal pumped at 1064 nm to be predicted accurately.
- Two methods for fine-scale wavelength tuning were investigated, crystal angle tuning and non-collinear angle tuning. In both cases good agreement was observed between measurement and theory.
- Theoretical calculations of the different acceptance angles were performed for potassium niobate and KTP, and the results compared against the values for lithium niobate. These calculations highlight the potential advantages of potassium niobate over lithium niobate.
- The conditions for polychromatic phase matching were investigated and found to be unobtainable in the required wavelength range.
- An alternative pumping scheme using 532 nm radiation was investigated. This offered much higher tuning rates, and narrow bandwidth operation was demonstrated over the entire wavelength range of interest with a single KTP crystal.

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8. APPENDIX 1 : VERIFICATION OF POLYCHROMATIC PHASE MATCHING

In this Appendix, we derive an analytical formula for polychromatic phase matching, as identified by Wang and Dunn⁹. This is the angle for which the variation of signal angle with wavelength passes through a minimum.

Assuming a fixed crystal angle θ , and applying the cosine rule to the vector triangle (Figure 9):

$$k_i^2 = k_p^2 + k_s^2 - 2 * k_p * k_s * \cos(\alpha) \quad (1)$$

Polychromatic phase matching occurs when k_s can change without changing k_p or α . Therefore, differentiate with respect to k_s and set $\delta k_p / \delta k_s$ and $\delta \alpha / \delta k_s$ to zero:

$$2 * k_i * \left(\frac{\partial k_i}{\partial k_s} \right) = 0 + 2 * k_s - 2 * k_p * \cos(\alpha) \quad (2)$$

Returning to the vector triangle (Figure 9) and resolving parallel to k_p

$$k_p = k_s * \cos(\alpha) + k_i * \cos(\beta) \quad (3)$$

Substituting this into (2) above:

$$\frac{\partial k_i}{\partial k_s} * k_i = k_s - k_s * \cos(\alpha)^2 - k_i * \cos(\beta) * \cos(\alpha) \quad (4)$$

Resolving perpendicular to k_p in the vector triangle

$$k_s * \sin(\alpha) = k_i * \sin(\beta)$$

Substituting this into (4)

$$r \frac{\partial k_i}{\partial k_p} * k_i = k_i * \sin(\alpha) * \sin(\beta) - k_i * \cos(\beta) * \cos(\alpha)$$

Hence, polychromatic phase matching occurs when

$$\frac{\partial k_i}{\partial k_s} = - \cos(\alpha + \beta) \quad (7)$$

Having calculated the conditions for polychromatic phase matching, we need to evaluate α and β individually. To do this, apply the sine rule to the vector triangle:

$$\frac{k_i}{\sin(\alpha)} = \frac{k_p}{\sin(180 - \alpha - \beta)} = \frac{-k_p}{\sin(\alpha + \beta)}$$

Hence

$$\sin(\alpha + \beta) = -\frac{k_p}{k_i} * \sin(\alpha) \quad (9)$$

If we now square (7) and add it to (9) and then re-arrange, we arrive at:

$$\sin^2(\alpha) = \frac{k_i^2}{k_p^2} * (1 - (\frac{\partial k_i}{\partial k_s})^2)$$

Equation 10 can then be used to determine α , and the result substituted in (9) to determine β .