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High-efficiency infrared generation by difference-frequency mixing using tangential phase matching

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Measurements have been made of the energy generated in the wavelength region 3.3 to 4.0 μm, by difference-frequency mixing between a tunable dye laser and a Nd:YAG laser in lithium niobate. The highest mixing efficiencies have been observed with the beams aligned in a non-collinear geometry. It is demonstrated that this non-collinear geometry corresponds to the tangential phase-matching condition which increases the acceptance angle of a second-order nonlinear interaction over that achieved in a non-critically phase-matched geometry.

1. Introduction

Difference-frequency mixing is an important technique for the generation of infrared radiation from visible and near-infrared laser sources. Particular examples include mixing between a *Q*-switched Nd:YAG laser (or its second harmonic) and a tunable dye laser in lithium niobate [1–3], lithium iodate [4,5] and in silver selenogallate [6]. However, the efficiency of difference-frequency mixing, and other second-order non-linear interactions, between non-diffraction limited beams is often limited by the acceptance angle of the interaction itself. This problem can sometimes be overcome by the use of non-critical phase matching [7]. However, non-critical (or 90°) phase matching is not applicable to all situations, so there is often a need for other phase-matching techniques that carry some of its advantages. One such technique is tangential phase matching [8], which increases the acceptance angle for one of the input waves by the use of a non-collinear geometry.

In this work tangential phase-matching has been demonstrated in a mixing experiment between a Nd:YAG laser (at 1.064 μm) and a dye laser (tunable between 785 and 851 nm) in lithium niobate. An energy conversion efficiency of 13% has been achieved resulting in the generation of energies of up to 12 mJ over the spectral region 3.3 to 4.0 μm. Preliminary investigations have shown the effective operation of this source at wavelengths up to 4.5 μm.

2. Theory of tangential phase matching

The frequencies of the waves involved in a difference-frequency mixing interaction are related by

$$\omega_h = \omega_f + \omega_g , \tag{1}$$

where $\omega_h > \omega_f > \omega_g$. The wavevector mismatch is defined as

$$\Delta \boldsymbol{k} = \boldsymbol{k}_f + \boldsymbol{k}_g - \boldsymbol{k}_h , \tag{2}$$

where $\Delta \boldsymbol{k}$ must be zero for the interaction to be perfectly phase matched. In the experiment performed here, ω_h and ω_g correspond to the frequencies of the tunable dye laser and the Nd:YAG laser respectively. The wave generated by the interaction is at the difference frequency ω_f. Since type I phase matching is used and the medium is negative uniaxial, ω_h must be an extraordinary wave and ω_f and ω_g must be ordinary waves.

The acceptance angle of such an interaction is determined by the sensitivity of eq. (2) to variations in the directions of the input waves. This sensitivity

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is closely linked to the geometry of the overlap between the loci of the wavevector surfaces. In a critically phase-matched interaction the phase fronts of the interacting waves intersect in the direction of propagation. Consequently the acceptance angle is quite small. It is often increased by the use of a phase-matched angle of 90° which ensures that the phase fronts of the input waves are tangential to the generated wave. This technique is known as non-critical phase matching [7,9]. The principle discussed here is tangential phase matching, which achieves a similar effect but by the use of a non-collinear geometry that enables the phase fronts of one of the input waves and the generated wave to be tangential. This principle has also been referred to as "one-beam non-critical" phase matching [10].

Fig. 1 shows a wavevector triangle for a second-order nonlinear process, such as difference-frequency mixing in which the wave propagate in a non-collinear geometry. The component of the phase mismatch (eq. (2)) parallel to the direction of propagation of the generated wave ($\boldsymbol{k}_\mathrm{f}$) is:

$$\Delta k_\ell = k_\mathrm{f} + k_\mathrm{g} \cos\phi - k_\mathrm{h}(\theta)\cos\gamma , \tag{3}$$

where $k_\mathrm{h}(\theta)$ is shown as a function of its direction θ because the case considered here it is an extraordinary wave. Since the Poynting vector is normal to the wavevector surface [11] the variation of the wavevector $k_\mathrm{h}(\theta)$ of the extraordinary wave with angle can be expressed as

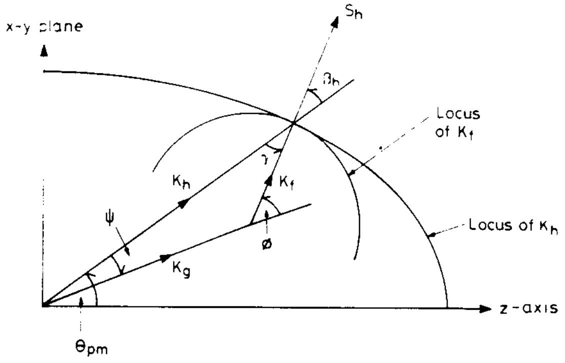

Fig. 1. Wave-vector triangle for difference-frequency mixing. The tangential phase-matching condition is achieved when the locus of $\boldsymbol{k}_\mathrm{h}$ is a tangent to the locus of $\boldsymbol{k}_\mathrm{f}$. $\boldsymbol{S}_\mathrm{h}$ is the Poynting vector of the extraordinary wave $\boldsymbol{k}_\mathrm{h}$, the angle β_h between $\boldsymbol{S}_\mathrm{h}$ and $\boldsymbol{k}_\mathrm{h}$ is the walkoff angle.

$$\partial k_\mathrm{h} / \partial\theta = -k_\mathrm{h} \tan\beta_\mathrm{h} , \tag{4}$$

where β is the walkoff angle given in terms of the ordinary and extraordinary refractive indices n_o and n_e as

$$\tan\beta = \frac{\sin\theta\cos\theta\,(n_\mathrm{o}^2 - n_\mathrm{e}^2)}{n_\mathrm{e}^2\cos^2\theta + n_\mathrm{o}^2\sin^2\theta} . \tag{5}$$

The sensitivity of eq. (3) to variations in the directions of the waves can be determined by differentiating it with respect to the angles shown in fig. 1. If the component of the phase mismatch perpendicular to the generated wave is negligible then the differential of the component of the phase mismatch parallel to the generated wave reduces to

$$\mathrm{d}(\Delta k_\ell) = k_\mathrm{g} \sin\phi\,\mathrm{d}\psi + k_\mathrm{h}\cos\gamma\,(\tan\gamma - \tan\beta_\mathrm{h})\,\mathrm{d}\theta . \tag{6}$$

This is an expression for the change in the phase mismatch in the direction of $\boldsymbol{k}_\mathrm{f}$ with respect to first-order changes in the direction of propagation of each of the interacting waves. In the derivation of this expression it has been assumed implicitly that the direction of each wave can vary independently of the others. It has been derived in a more general form elsewhere [12].

Eq. (6) shows that when $\gamma = \beta_\mathrm{h}$ the parallel component of the phase mismatch is insensitive to changes in θ, and therefore is also insensitive to changes in the direction of $\boldsymbol{k}_\mathrm{h}$. If the interaction is close to being phase matched ($\Delta\boldsymbol{k} = 0$ in eq. (2)) then this is achieved when

$$\psi = \sin^{-1}\left(\frac{k_\mathrm{f}}{k_\mathrm{g}} \sin\beta_\mathrm{h}\right) . \tag{7}$$

This is the tangential phase-matching condition. It has been referred to by previous authors [8,13] but has never been expressed in this concise form before. In the experiment described below, the walkoff angle β_h calculated from eq. (5) is 2.0°. Hence, if the generated wave is at 3.59 μm, $\psi = 0.6°$ calculated internally to the crystal. The corresponding external angle is 1.3° for a lithium niobate crystal (assuming $n_\mathrm{e} = 2.2$).

3. Experiment

3.1. Measurement of phase-matching angles

The application of the tangential phase-matching technique to the generation of near-infrared radiation by difference-frequency mixing has been demonstrated using the apparatus shown in fig. 2. A pulsed *Q*-switched Nd:YAG laser (Quantel International YG682) was used as one of the inputs for the experiment together with a tunable dye laser (Quantel International TDL 60) pumped by the second harmonic of the Nd:YAG laser. The Nd:YAG laser was injection seeded and had a linewidth of 0.002 cm^{-1} and a pulse length of 6 to 8 ns. The dye laser used LDS 821 dye (Exciton) in methanol and was tunable over the wavelength range 785–851 nm with a linewidth of 0.1 cm^{-1}.

The difference-frequency mixing was performed in a lithium niobate crystal with dimensions $50\times 11\times 11$ mm^3 which was held in a temperature-stabilised oven. It was orientated with its principal plane vertical and was cut at a nominal angle of 47° to the optic axis. The input Nd:YAG and tunable dye laser beams were polarised horizontally and vertically respectively. A rotating-plate attenuator (Newport Research Corp) was used to vary the energy from the tunable dye laser incident on the mixing crystal.

A practical advantage resulting from the use of a non-collinear geometry is that the dichroic beam-combiner BS2 (in fig. 2) can be replaced with a simple reflector split in half along a diameter. Similarly, at the output from the mixing crystal, the beams are propagating in different directions and are therefore easy to separate.

Initially, an experiment was performed to establish the angles between the input beams that resulted in the generation of the highest energy at the difference frequency. The angles that gave the highest mixing energy are shown in fig. 3. The theory outlined above suggests that the acceptance angle of the interaction will be a maximum when the tangential phase-matching condition is met. However, this theory does not allow either for the effect of beam overlap within the crystal or for the non-uniform profile of the beams. The first effect results in a decrease in the energy generated, because of the reduction in the length of the interaction region within the crystal as the angle between the beams increases. The second effect is more difficult to quantify and could result in an increase or a decrease in the angle. In this experiment an optimum external angle of 17 mrad (=0.97°) was measured. This is slightly less than the angle calculated in the previous section, which suggests that there is an effect due to the overlap of

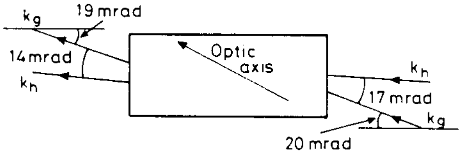

Fig. 3. Angles measured in the principal plane for optimum difference-frequency mixing. The positions of the beams at the input and output are affected by both refraction and Poynting vector walkoff and are not shown accurately in this figure.

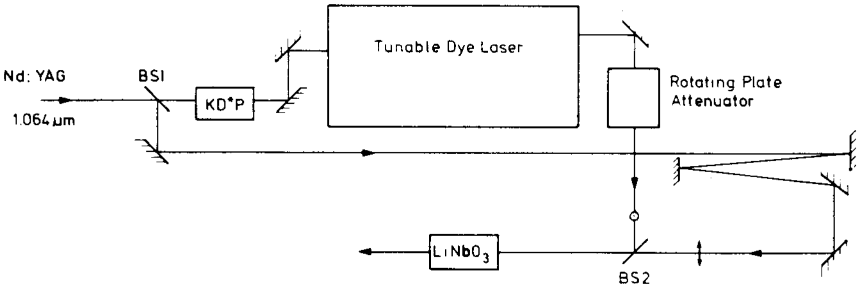

Fig. 2. Apparatus used for difference-frequency mixing. The dye laser beam is polarised vertically and the Nd:YAG laser beam is polarised horizontally at the mixing crystal. BS1 splits 30% of the Nd:YAG beam off for the mixing experiment. The remaining 70% is frequency doubled in KD*P (deuterated potassium di-hydrogen phosphate) to generate approximately 500 mJ of radiation to pump the dye laser. BS2 is a dichroic beam-combiner, but could be replaced by a split ("half moon") reflector.

the beams. However, the agreement is reasonable in view of the errors involved in measuring the angles.

Examples have been reported [14] where the optimum geometry is a mirror image of the one used here, with the ordinarily polarised input being directed in the walkoff-direction of the extraordinarily polarised input in order to maximise the length of their interaction within the crystal. This will produce optimum results when the overlap factor is dominant over the effect of the acceptance angle.

3.2. *Mixing efficiency*

The energy generated by the difference-frequency interaction is shown in figs. 4 and 5. In both of these graphs the energy generated at the difference frequency is linearly dependent on the energy from the tunable dye laser. Consequently, the performance of the interaction can be characterised by the efficiency defined as the quotient of the energy generated at difference frequency and the input energy at tunable dye laser frequency.

The results shown in fig. 4 correspond to an efficiency of 9.3% with an incident energy of 48 mJ at 1.064 µm. This corresponds to a photon conversion efficiency of 41%. The efficiency is not significantly altered by reducing the 1.064 µm input to 28.5 mJ because the efficiency is limited by the number of photons available from the dye laser. Fig. 5 shows the energy generated at the difference frequency together with the energy measured at 1.064 µm at the output of the crystal. This beam is the lower frequency input beam (ω_g in eq. (1)) and is therefore amplified by the interaction. The parametric gain observed is comparable to that observed in other experiments at similar wavelengths [15].

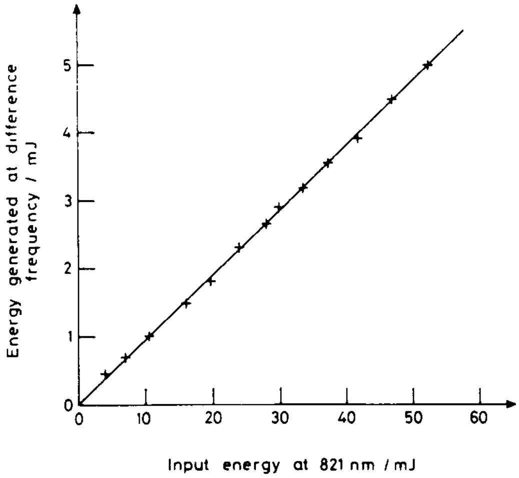

Fig. 4. Energy generated at the difference frequency (3.59 µm) versus input energy at 821 nm, for inputs of 48 and 28.5 mJ at 1.064 µm.

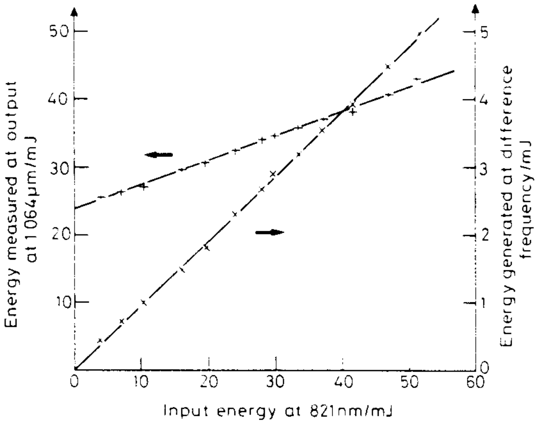

Fig. 5. Energy generated at the difference frequency (3.59 µm) and amplified output at 1.064 µm versus input energy at 821 nm, for an input of 30 mJ at 1.064 µm.

In order to maximise the energy available from the dye laser the rotating-plate attenuator was removed. This resulted in an energy of 7.5 mJ at 3.59 µm being generated from 30 mJ at 1.064 µm and 76 mJ at 821 nm. This corresponds to an efficiency of 9.9% (or a photon conversion efficiency of 43.3%).

In these experiments the mixing crystal was approximately 1.5 m from the exit of the dye laser. In order to perform the same experiment with a smaller diameter dye laser beam in the crystal, this distance was reduced while retaining the same angle between the beams. For practical reasons, this required both polarisations to be rotated through 90° and the lithium niobate to be orientated with its principal plane horizontal. This was achieved using a tunable polarisation rotator (Newport Research Corp) in the dye laser beam and a half-wave plate in the 1.064 µm beam. These modifications enabled the experiment to be performed 55 cm from the exit of the dye laser using a split reflector in place of BS2. Using a fresh dye solution an energy of 12 mJ at 3.59 µm was generated from 92 mJ at 821 nm and 115 mJ at 1.064 μm. This represents a conversion efficiency of 13% or a photon conversion efficiency of 56%. When the 1.064 μm beam was spatially filtered using an apodizer its energy was reduced by approximately 50%, yet there was only a small reduction in the energy generated at 3.59 μm (to 10.5 mJ). This suggests that only 50% of the Nd:YAG laser beam fell within a diffraction-limited cone.

3.3. Tuning range

The measurements described above were all made using the dye LDS 821, which has a nominal tuning range of 785 to 851 nm. An investigation of the mixing efficiency over this tuning range showed that an efficiency of at least 10% could be achieved over the range 800 to 825 nm, which corresponds to a range of 3.3 to 4.0 μm for the generated radiation. Using the dye LDS 867 measurements were made at longer wavelengths. As the wavelength was increased the efficiency decreased to 6% at a wavelength of 861 nm, corresponding to 4.5 μm. This wavelength is very close to the expected cut-off wavelength of lithium niobate. There is no reason to believe that there is any other limit on the shortest wavelength generated, apart from the tuning range of the dye laser.

4. Conclusions

The principle of tangential phase matching has been established theoretically and demonstrated experimentally. Its use allows the acceptance angle of a non-linear interaction to be increased by using a non-collinear alignment of the interacting beams. This principle should be applicable to a wide range of other second-order non-linear interactions.

Tunable difference-frequency mixing has been demonstrated with an efficiency of 13% using the tangential phase-matching technique. This efficiency remains nearly constant over the tuning range 3.3 to 4.0 μm. This range has been extended to 4.5 μm with only a slight reduction in efficiency.

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References

[1] K. Kato, IEEE J. Quantum Electron. 16 (1980) 1017.
[2] J.J. Jacob, SPIE 461 (1984) 11.
[3] D.S. Bethune and A.C. Luntz, Appl. Phys. B 40 (1986) 107.
[4] L. Mannik and S.K. Brown, Optics Comm. 57 (1986) 360.
[5] K. Kato, IEEE J. Quantum Electron. 21 (1985) 119.
[6] K. Kato, IEEE J. Quantum Electron. 20 (1984) 698.
[7] F. Zernike and J. Midwinter, Applied nonlinear optics (Wiley, New York, 1973).
[8] N.P. Barnes and V.J. Corcoran, Appl. Optics 15 (1976) 696.
[9] R.C. Miller, G.D. Boyd and A. Savage, Appl. Phys. Lett. 4 (1965) 77.
[10] S.X. Dou, D. Josse and J. Zyss, J. Opt. Soc. Am. B., to be published.
[11] H. Kogelnik and H. Motz, Symposium on Electromagnetic Theory and Antennas (Pergamon Press, New York, 1963) p. 477.
[12] M.J.T. Milton, Ph.D. Thesis, Southampton University (1990).
[13] J. Warner, Opto-Electron. 1 (1969) 25.
[14] H. Gerlach, Optics Comm. 12 (1974) 405.
[15] M.J.T. Milton and T.J. McIlveen, Optics Comm. 82 (1991) 153.