Broadband pump wavelength tuning of a low threshold *N*-(4-nitrophenyl)-*L* prolinol near infrared optical parametric oscillator

S. Khodja, D. Josse, I. D. W. Samuel, and J. Zyss
France Telecom, CNET-Paris B, Laboratoire de Bagneux, Départment d'Electronique Quantique et Moléculaire, 196 Avenue Henri Ravéra, BP107, 92225 Bagneux Cedex, France

(Received 17 July 1995; accepted for publication 11 October 1995)

We report the pump wavelength tuning of a low threshold *N*-(4-nitrophenyl)-*L*-prolinol based nanosecond pulsed optical parametric oscillator in the near infrared. A broad tuning range from 1 to 1.5 μm corresponds to a limited pump wavelength excursion from 583 to 590 nm and a record low oscillation threshold of the order of 0.5 MW cm^{-2}. © *1995 American Institute of Physics.*

Rapidly tunable coherent sources of near-infrared radiation are required for high data rate wavelength division multiplexing (WDM) optical communication networks, photonic switching systems, and related optical signal processing applications. Optical parametric oscillators (OPOs) are promising in this perspective, whereas the use of the organic crystal *N*-(4-nitrophenyl)-*L* prolinol (NPP)[1] enables one to demonstrate very low oscillation thresholds.[2–4]

In most of the previous investigations, the tunability of the output wavelength of OPOs is ensured by either angle or thermal tuning, which limits the wavelength switching dynamics due to mechanical or thermal inertia constraints. Pump wavelength tuning in $KTiOPO_4$ (KTP) crystals has been demonstrated by use of a type II phase-matching configuration. The ratio of signal to idler wavelengths tuning range over that of the pump ($\Delta\lambda_{i,s}/\Delta\lambda_p$) was, however, limited to 5.[5]

In this letter we report the demonstration of broadband pump wavelength tuning of an NPP-based OPO with a signal to idler wavelength tuning ratio over 100. Tuning by the pump offers the prospect of wavelength switching speeds many orders of magnitude larger than achievable by angle or thermal tuning.

The output wavelengths of the OPO (signal and idler) are determined by the two well-known conditions for energy and momentum conservation:

$$\omega_p = \omega_i + \omega_s, \tag{1a}$$

$$\mathbf{K}_p = \mathbf{K}_i + \mathbf{K}_s, \tag{1b}$$

where $\mathbf{K}$ is the wave vector and subscripts p, i, and s refer, respectively, to the pump, idler, and signal beams.

In addition to the more traditional ways (using the angle dependence of the material birefringence or by thermal effects), the dispersion of the index of the nonlinear material can be taken advantage of by varying the pump wavelength. In order to be useful for applications, this effect must ensure an OPO output tunability at least one order of magnitude larger than the corresponding excursion of the pump wavelength.

The wave-vector mismatch in parametric interactions is

$$\mathbf{\Delta K} = \mathbf{K}_p - \mathbf{K}_i - \mathbf{K}_s, \tag{2}$$

where the wave vector is given by

$$K_j = n(\omega_j)\omega_j/c, \quad j = p, i, \text{ and } s, \tag{3}$$

c being the speed of light in vacuum.

We consider a type I phase-matching configuration[2,3] corresponding to the actual experimental situation in NPP, whereby the pump is polarized along the Y axis and the signal and idler are polarized in the (XZ) plane. In the colinear case, the phase mismatch vector can be expressed as

$$\Delta K(\omega_p, \omega_i, \omega_s) = [\omega_p n_0(\omega_p) - \omega_i n_e(\omega_i) - \omega_s n_e(\omega_s)]/c, \tag{4}$$

where the ordinary refractive index along the Y axis at pump frequency is given by

$$n_0(\omega_p) = n_y(\omega_p) \tag{5}$$

and the extraordinary refractive index in the XZ plane at idler and signal frequencies is given by

$$n_e(\omega_{i,s}) = \left(\frac{\sin^2(\theta)}{n_z^2(\omega_{i,s})} + \frac{\cos^2(\theta)}{n_x^2(\omega_{i,s})} \right)^{-1/2}, \tag{6}$$

where θ is the angle between the direction of propagation of the light beam (wave vector) and the Z axis in the (XZ) plane.

When the phase-matching condition is satisfied at the operating point $\omega_p = \omega_{p0}$, conditions (1a) and (4) lead to

$$\omega_{p0} = \omega_{i0} + \omega_{s0}, \tag{7}$$

$$\omega_{p0} n_0(\omega_{p0}) - \omega_{i0} n_e(\omega_{i0}) - \omega_{s0} n_e(\omega_{s0}) = 0. \tag{8}$$

Differentiating Eq. (4) at the vicinity of the central operating pump wavelength ω_{p0} (corresponding to the emission wavelengths ω_{i0} and ω_{s0}) for a fixed phase-matching angle θ leads to the following relationship between $\Delta\omega_i$ and $\Delta\omega_p$:

$$\Delta\omega_p(\Delta K'_{\omega_p}|_0 + \Delta K'_{\omega_s}|_0) + \Delta\omega_i(\Delta K'_{\omega_i}|_0 - \Delta K'_{\omega_s}|_0) = 0, \tag{9}$$

where we have used

$$\Delta\omega_s = \Delta\omega_p - \Delta\omega_i, \tag{10}$$

and to differentiate from Eq. 1(a) $\Delta K'_{\omega_p}|_0$ refers to the partial derivative of ΔK with respect to ω_p taken at the central operating wavelengths (similar notations are used for $\Delta K'_{\omega_i}$ and $\Delta K'_{\omega_s}$).

The partial derivative of the mismatch wave vector with respect to the pump frequency at ω_{p0} can be expressed as:

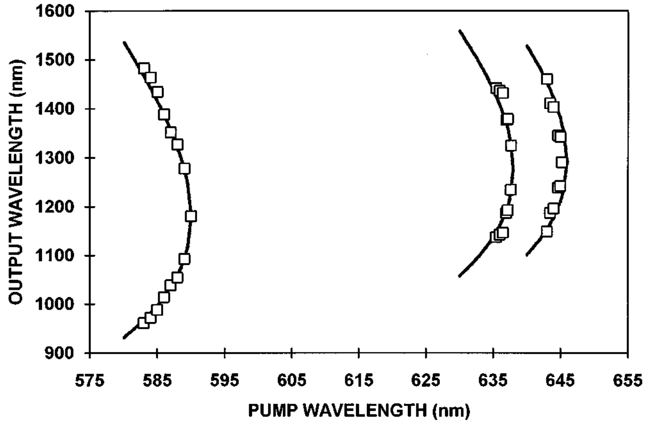

FIG. 1. NPP-based OPO: Tuning of the output signal and idler wavelengths by variation of the pump wavelength: the solid line is from calculation, and the square points are experimental. The three curves from left to right correspond to phase-matching angles of 17°, 30°, and 32°, respectively.

$$\Delta K'_{\omega_p}|_{\omega p0} = \frac{1}{c}\left(n_0(\omega_{p0}) + \omega_{p0} \frac{\partial n_0(\omega)}{\partial \omega}\bigg|_{\omega p0} \right). \tag{11}$$

Group velocities of the interacting waves can be conveniently introduced in Eq. (9) with:

$$\Delta K'_{\omega_p}|_0 = \frac{1}{v_g(\omega_{p0})}, \tag{12}$$

$$\Delta K'_{\omega_i,\omega_s}|_0 = -\frac{1}{v_g(\omega_{i0},\omega_{s0})}, \tag{13}$$

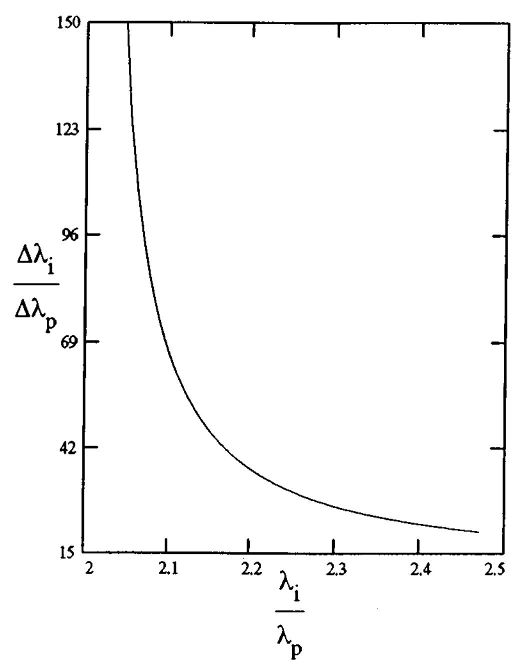

FIG. 2. Ratio of the idler wavelength variation of the pump wavelength range, $\Delta\lambda_i/\Delta\lambda_p$, with respect to the idler to pump wavelength ratio λ_i/λ_p.

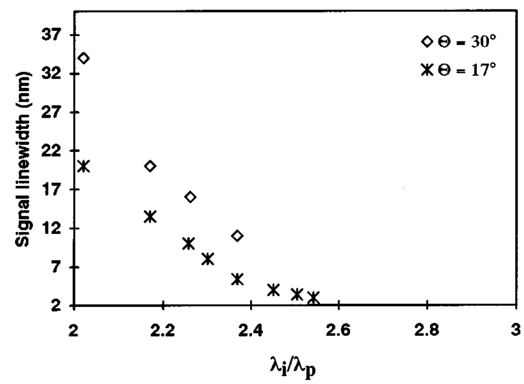

FIG. 3. Experimental measurement of the signal linewidth of the NPP-based OPO tuned by pump wavelength variation as a function of the λ_i/λ_p idler over pump wavelength ratio for two different external angles: $\theta=17°$ corresponds to a pump wavelength λ_p of 590 nm at degeneracy; and $\theta=30°$ corresponds to a pump wavelength λ_p of 646 nm at degeneracy.

where $v_g(\omega_{p0})$, $v_g(\omega_{i0})$, and $v_g(\omega_{s0})$ refer to the group velocities, respectively, at pump, idler, and signal frequencies.

Equation (9) finally takes the form

$$\frac{\Delta\omega_i}{\Delta\omega_p} = \frac{\beta_{ps}}{\beta_{is}}, \tag{14}$$

where

$$\beta_{ij} = \frac{1}{v_g(\omega_{i0})} - \frac{1}{v_g(\omega_{j0})}.$$

The ratio reflects, the sensitivity of the emitted wavelength with respect to the pump wavelength detuning away from an initial configuration corresponding to the interacting wavelengths ω_{p0}, ω_{i0}, and ω_{s0}.

We note from Eq. (14) that when the OPO output wavelength is close to degeneracy, the signal and idler take identical values $[v_g(\omega_{i0}) \approx v_g(\omega_{s0})]$ leading to divergence of the $\Delta\omega_i/\Delta\omega_p$ ratio, and the well-known linewidth broadening of the OPO output. On the contrary, when the OPO output wavelength is far off degeneracy, the difference between the idler and signal group velocities $v_g(\omega_{i0}) - v_g(\omega_{s0})$ tends to increase and, consequently, the value of the factor $\Delta\omega_i/\Delta\omega_p$ ratio tends to decrease.

It is worth noting that at the group velocity matching configuration $[v_g(\omega_{i0}) = v_g(\omega_{s0})]$ that may occur at degen-

TABLE I. Spectral features of the NPP-based OPO tuned by pump wavelength variation at the point $\lambda_i/\lambda_p = 2.27$.

θ (°)	λ_p at degeneracy (nm)	$\Delta\lambda_i/\Delta\lambda_p$	Signal linewidth (nm)	Threshold MW cm^{-2}
17	590	30	8	0.45
30	538	35	16	0.45
32	646	40	17	0.45

eracy in type I phase matching, the expansion of ΔK in Eq. (9) must include second-order terms that take into account group velocity dispersion.

In this case, the rate of change in the output wavelength versus the change in the pump wavelength is given by the following expression:

$$\Delta\omega_p\beta_{ps}-\Delta\omega_i\beta_{is}+\tfrac{1}{2}(\Delta\omega_p)^2\gamma_{ps}-\tfrac{1}{2}(\Delta\omega_i)^2(\gamma_i+\gamma_s)+\Delta\omega_p\Delta\omega_i\gamma_s=0, \tag{15}$$

where

$$\gamma_{ij}=\frac{\partial}{\partial\omega_i}\left(\frac{1}{v_g(\omega_{i0})}\right)-\frac{\partial}{\partial\omega_j}\left(\frac{1}{v_g(\omega_{j0})}\right),$$

$$\gamma_i=\frac{\partial}{\partial\omega_i}\left(\frac{1}{v_g(\omega_{i0})}\right).$$

For group velocity matching at degeneracy $\beta_{is}=0$ and Eq. (14) becomes

$$\Delta\omega_p\beta_{ps}+(\Delta\omega_p)^2\gamma_{ps}-(\Delta\omega_i)^2(\gamma_i+\gamma_s)+2\Delta\omega_p\Delta\omega_i\gamma_s=0. \tag{16}$$

This equation can be used together with Eq. (14) to trance numerically the dependency of the emitted wavelength with respect to the pump wavelength variation.

Measurements were performed in three series of experiments, with BBO-based OPO pumped by the third harmonic of a Q-switched Nd^{3+}:YAG laser as a tunable pump source (pulse duration 7 ns). The linewidth of the OPO in the spectral region from 580 to 650 nm varies slowly and is around 3 nm. A type I phase-matching configuration of NPP[6] was used, with the pump polarized along the Y axis, and the signal and idler polarized in the (XZ) plane.

In the first series of experiments, the singly resonant OPO cavity consisted of two mirrors that were highly reflective at the signal wavelength and transmitting at the pump wavelength. The input mirror was curved with a radius of 500 mm while the output mirror was planar, the cavity being 14 mm long. The angle of the crystal θ was 17° with respect to the normal to the as-grown (101) entrance surface of the NPP crystal, corresponding to degenerate phase matching for a pump excitation at 590 nm.

As the pump wavelength was tuned from 583 to 590 nm, the signal and idler wavelengths varied from 961 to 1482 nm: a change of 1 nm of the pump wavelength thus leads to an excursion of the output wavelength by more than 70 nm. The results, as shown in Fig. 1, are in good agreement with numerical calculations based on Eqs. (14) and (16).

In Fig. 2 we report the $\Delta\lambda_i/\Delta\lambda_p$ ratio of the idler wavelength change with respect to that of the pump as a function of the idler to the pump wavelength ratio λ_i/λ_p. This factor referred to in Kovrigin and Byer[7] as a stability factor can be varied in our case from 40 when the OPO wavelength is far away from degeneracy (at $\lambda_i=1482$ nm and $\lambda_p=583$ nm) to several hundred when the OPO wavelength output is close to degeneracy.

The linewidth of the OPO increases with that of the $\Delta\lambda_i/\Delta\lambda_p$ factor and becomes very large. This feature explains in part the large linewidth of the OPO close to degeneracy corresponding to an infinite value of the $\Delta\lambda_i/\Delta\lambda_p$ factor. Signal linewidth measurements are presented in Fig. 3. The signal linewidth varied from 3 nm at $\lambda_s=961$ nm and $\lambda_p=583$ nm to more than 16 nm close to degeneracy.

In the two other series of experiments, the OPO resonator was formed by two mirrors that were highly reflective in the region of 1300 nm, and highly transmitting at the pump wavelength. The input coupler mirror has a curvature radius of 3000 mm, the output mirror being planar and the cavity 15 mm long.

We investigated the OPO output wavelength tuning by pump wavelength variation for pump wavelengths shifted toward the red range at different phase-matching angles: for a phase-matching angle θ of 30°, when the pump was tuned from 635 to 638 nm, the OPO output wavelength excursion extended from 1137 to 1439 nm. Furthermore, for a phase matching angle θ of 32°, tuning the pump from 643 to 646 nm leads to an OPO output extending from 1148 to 1460 nm (see Fig. 1). In these two experiments, the ratio of signal-idler wavelength tuning range over that of the pump raises to more than 100. It should be noted that in this series of experiments, the oscillation threshold remains remarkably low, that is, of the order of 0.5 MW/cm^2, close to degeneracy in conditions that would require tens of MWs in classically used inorganic crystal-based OPOs.

In conclusion, we have demonstrated, in an organic material, the tuning of the output of an OPO by variation of the pump wavelength. The change in output wavelength can be more than 100 times larger than the change in the pump wavelength. Pump wavelength tuning provides advantages over already existing tuning schemes, opening up perspectives at high wavelength multiplexing rates. The noncriticality of the λ_s vs λ_p relationship can be of interest in femtosecond applications.[6]

[1] See, for example, *Molecular Nonlinear Optics: Materials, Physics, and Devices*, edited by J. Zyss (Academic, Boston, 1993); J. Zyss, J. F. Nicoud, and M. Coquillay, J. Chem. Phys. **81**, 4160 (1984); J. Zyss, J. Phys. D **26**, B198 (1993).
[2] D. Josse, S. X. Dou, and J. Zyss, Appl. Phys. Lett. **61**, 121 (1992).
[3] S. X. Dou, D. Josse, and J. Zyss, J. Opt. Soc. Am. B **10**, 1708 (1993).
[4] D. Josse, S. Khodja, J. Badan, I. D. W. Samuel, and J. Zyss, Appl. Phys. Lett. **64**, 3655 (1994).
[5] M. G. Jani, R. C. Powell, B. Jassemnejad, and R. Stolzenberger, Appl. Opt. **31**, 1998 (1992).
[6] I. Ledoux, C. Lepers, A. Périgaud, J. Badan, and J. Zyss, J. Opt. Soc. Am. B **4**, 987 (1987).
[7] A. I. Kovrigin and R. Byer, IEEE J. Quantum Electron. **5**, 384 (1969).