Optical Parametric Amplification

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Abstract—This paper presents a detailed theoretical and experimental study of optical parametric amplification. Experimental measurements in KD*P pumped at 354.7 nm and LiNbO₃ pumped at 1064 nm by a Q-switched Nd:YAG source are in excellent agreement with theory. The large gains, high conversion efficiency, and wide tuning range make optical parametric amplification an important method for power generation in the optical and infrared spectral range.

I. INTRODUCTION

Experiments in nonlinear optics were performed in 1961 soon after the demonstration of the laser [1]. These experiments were possible due to the increase in power spectral brightness made possible by the laser. The first mixing experiment involving three optical frequencies was performed by Wang and Racette [2] in 1965. Prior to that experiment the possibility of parametric gain in a three frequency process was considered theoretically by Kingston [3], Kroll [4], Akhanov and Khoklov [5], and Armstrong, Bloembergen, Ducuing, and Pershan (ABDP) [6].

Parametric oscillation, an important extension of parametric amplification, was first achieved in 1965 by Giordimaine and Miller [7]. Parametric oscillation is quite useful for generation of widely tunable coherent radiation. The progress in parametric amplification and oscillation has been the subject of review papers by Harris [8], Smith [9], and Byer [10].

Since the original experiment of Wang and Racette, laser sources for pumping parametric processes have improved considerably. Pulsed lasers are now available with energies greater than 0.5 J/pulse at repetition rates greater than ten pulses per second and operating with either a TEM₀₀ transverse mode profile or various plane wave unstable resonator beam outputs. Recent LiNbO₃ parametric amplification experiments demonstrated that small-signal gains of 50 and saturated conversion efficiencies of 20 percent are available. Consequently, parametric amplification is an important technique for amplification and power generation over wide frequency ranges. The detailed understanding of the parametric amplification process is also useful for the design of optical parametric oscillator tunable sources.

This paper examines the theoretical solutions of parametric amplification valid in the depleted pump regime. The solutions are compared with experimental measurements for KD*P and LiNbO₃ optical parametric amplifiers (OPA). The theoretical derivations, based on previous work by ABDP [6], Boyd and Kleinman (BK) [11], and Harris [12] and computational methods are considered in Sections II and III. The theoretical treatment includes the transition from parametric amplification solutions with pump depletion to solutions valid where pump depletion is not important. A parametric amplifier general solution program with and without time dependence is discussed in Section III.

The experimental measurements are presented in Section IV. The experiments include small-signal gain measurements of a 355 nm pumped KD*P OPA and its angle tuning curve. The gain and energy conversion efficiency of a 1064 nm pumped LiNbO₃ OPA is also presented. The experimental results are compared with the computer solutions presented in Section III.

The theoretical and experimental results and comparisons presented in this paper are the first detailed study of the optical parametric amplifier. Together they show that OPA's are useful devices for amplification of tunable coherent radiation in the optical spectral region.

II. THEORY

A. Second-Order Nonlinear Interactions

The second-order nonlinear interaction of light beams is characterized by the generation of a nonlinear polarization

$$\vec{P}_3(\omega_3) = \varepsilon_0 \vec{X}(-\omega_3, \omega_1, \omega_2): \vec{E}_1(\omega_1)\vec{E}_2(\omega_2)$$

(1)

where $\vec{P}_3(\omega_3)$ is the nonlinear polarization at frequency $\omega_3$, $\varepsilon_0$ is the dielectric constant, $\vec{X}(-\omega_3, \omega_1, \omega_2)$ is the nonlinear susceptibility tensor, and $\vec{E}_1(\omega_1)$ and $\vec{E}_2(\omega_2)$ are the interacting laser fields.

The second-order nonlinear susceptibility is responsible for sum and difference frequency generation and for parametric amplification in the interaction of two laser fields in a nonlinear medium. Parametric amplification involves three frequencies related by

$$\omega_3 = \omega_1 + \omega_2.$$  

(2)

The field at $\omega_3$ is assumed to be the most intense. Energy at either $\omega_1$ or $\omega_2$ incident on the nonlinear crystal is amplified with the remaining nonincident field being generated. For an efficient transfer of energy to occur between the waves, momentum or phase velocity phase matching, defined by the $k$ vector relation

$$k_3 = k_1 + k_2$$

(3)

must be accomplished, where $|k_i| = 2\pi n_i/\lambda_i$ and $n_i$ is the index of refraction in the crystal. For uniaxial crystalline media with birefringence, phase velocity matching [13] can be
achieved by angle tuning, crystal heating, or electrooptic effect. In the present experimental work, Type I angle phase matching was used with the signal (\(a_1\)) and idler (\(a_2\)) fields polarized as ordinary and the pump (\(a_3\)) field polarized as extraordinary waves in the negative birefringent LiNbO_3 and KD*P crystals. In negative uniaxial crystals the Type I phase-matching condition for collinear propagating waves is given by

\[
\lambda_3/n_\theta(\lambda_3, \theta) = \lambda_2/n_\theta(\lambda_2) + \lambda_1/n_\theta(\lambda_1)
\]

where

\[
\left[ \frac{1}{n_\theta(\lambda_3, \theta)} \right]^2 = \frac{\sin^2 \theta + \cos^2 \theta}{n_\theta^2(\lambda_3) + n_\theta^2(\lambda_3)}.
\]

(4)

Here \(\theta\) is the propagation angle relative to the optic axis and \(n_\theta(\lambda)\) and \(n_\theta(\lambda)\) the principle extraordinary and ordinary crystalline indexes of refraction.

A phase velocity mismatch factor

\[
\Delta k = k_1 - k_2 - k_3
\]

is a modification of (3) when phase matching is not quite achieved. Possible causes of nonzero \(\Delta k\) as well as finite crystal acceptance angle and bandwidth, are discussed in Appendix I.

The pump beam propagating as an extraordinary ray in the nonlinear crystal has a power flow direction at an angle \(\beta\) to the wavefront normal. The Poynting vector walkoff angle for uniaxial crystals is given by

\[
\tan \beta = \frac{\sin \theta_m \cos \theta_m (n_m^2 - n_\theta^2)}{n_m^2 \cos^2 \theta_m + n_\theta^2 \sin^2 \theta_m}
\]

with a positive angle \(\beta\) for positive birefringent crystals. The presence of \(\beta\) causes a slight adjustment in the observed value of the phase-matching angle \(\theta_m\) given by

\[
\theta_m_{\text{obs}} = \theta_m_{\text{calc}} + \beta.
\]

The specific nonlinear polarization relations derived from (1) are

\[
P_1 = 2\varepsilon_0 d_{\text{eff}} E_1^2 E_3
\]

\[
P_2 = 2\varepsilon_0 d_{\text{eff}} E_1^2 E_3
\]

\[
P_3 = 2\varepsilon_0 d_{\text{eff}} E_1 E_2
\]

where the notation has been simplified by dropping the explicit frequency dependence and the susceptibility has been written in terms of an effective nonlinear coefficient. For LiNbO_3 with 3\(m\) point group symmetry

\[
d_{\text{eff}} = d_{31} \sin \theta_m - d_{22} \cos \theta_m \sin 3\phi
\]

which is maximized for \(\phi = -90^\circ\). For KD*P with 42\(m\) point group symmetry the effective nonlinear coefficient for Type I phase matching is

\[
d_{\text{eff}} = -d_{41} \sin \theta_m \sin 2\phi
\]

which is maximized for \(\phi = -45^\circ\). A derivation of the effective nonlinear coefficient used here is presented in Appendix 2 of BK [11]. The coordinate rotations necessary to relate \(d_{\text{eff}}\) to the \(d_{\text{HT}}\) coefficient along the crystalogaphic axes are discussed in Zernike and Midwinter [14].

B. Coupled Wave Equations

The derivation of the coupled set of wave equations proceeds from Maxwell's equations

\[
\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}
\]

\[
\nabla \times \vec{B} = \vec{J} + \frac{\partial \vec{D}}{\partial t}
\]

(11a)

\[
\nabla \cdot \vec{D} = 0
\]

\[
\nabla \cdot \vec{B} = 0
\]

(11b)

and the constitutive relations

\[
\vec{D} = \varepsilon_0 \varepsilon_\infty \vec{E} + \vec{P}
\]

(12a)

\[
\vec{B} = \mu_0 \vec{H}
\]

(12b)

The linear polarization is contained in \(\vec{e}\) so that \(\vec{P}\) contains only the nonlinear polarization terms. The medium is assumed to be magnetically inactive. The fields obey the wave equation

\[
\frac{\partial^2 \vec{E}}{\partial t^2} - \mu_0 \sigma \frac{\partial \vec{E}}{\partial t} - \mu_0 \varepsilon_0 \varepsilon_\infty \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}
\]

(13)

and are assumed to be monochromatic plane waves propagating in the near field in the \(z\) direction. The nonlinear interaction occurs over distances and times that are large with respect to the individual sinusoidal variations of optical fields so that an envelope representation

\[
E_1(x, t) = \sum_m R_m(E_m \exp j(\omega_m t - k_m z))
\]

(14a)

\[
P_1(x, t) = \sum_m R_m(P_m \exp j(\omega_m t - k_m z))
\]

(14b)

is used. The subscript \(m\) represents the direction of polarization \(x\) or \(y\) and \(m\) the frequency of interest 1, 2, or 3. Equations (14a) and (14b) are substituted into (13) and the slowly varying envelope approximations are applied leading to

\[
\frac{\partial E_m}{\partial z} + (n_m/c) \frac{\partial E_m}{\partial t} + \alpha_m E_m = -j \frac{\mu_0 \varepsilon_\infty \omega_m}{2n_m} P_m
\]

(15)

where \(n_m = (\varepsilon_m)^{1/2}\) and \(\alpha_m = \frac{1}{2} \mu_0 \varepsilon_0 \varepsilon_\infty n_m\) is the electric field loss factor. The slowly varying envelope approximation is quite valid at the wavelengths, intensities, and pulse lengths relevant to the present experimental investigations. The envelope representation applied to the nonlinear polarization components given by (8) gives

\[
P_1 = 2\varepsilon_0 d_{\text{eff}} E_1^2 E_3 e^{-j\Delta k z}
\]

(16a)

\[
P_2 = 2\varepsilon_0 d_{\text{eff}} E_1^2 E_3 e^{-j\Delta k z}
\]

(16b)

\[
P_3 = 2\varepsilon_0 d_{\text{eff}} E_1 E_2 e^{j\Delta k z}
\]

(16c)

These polarization components substituted into (15) produce the coupled set of equations

\[
\frac{\partial E_1}{\partial z} + \frac{n_1}{c} \frac{\partial E_1}{\partial t} + \alpha_1 E_1 = -j \frac{\omega_1 d_{\text{eff}}}{n_1 c} E_3^* E_3 e^{-j\Delta k z}
\]

(17a)

\[
\frac{\partial E_2}{\partial z} + \frac{n_2}{c} \frac{\partial E_2}{\partial t} + \alpha_2 E_2 = -j \frac{\omega_2 d_{\text{eff}}}{n_2 c} E_3^* E_3 e^{-j\Delta k z}
\]

(17b)

\[
\frac{\partial E_3}{\partial z} + \frac{n_3}{c} \frac{\partial E_3}{\partial t} + \alpha_3 E_3 = -j \frac{\omega_3 d_{\text{eff}}}{n_3 \cos^2 \beta} E_1 E_2 e^{j\Delta k z}
\]

(17c)
The experiments operate in frequency regions where crystal losses are negligible, i.e., $\alpha_1 = \alpha_2 = \alpha_3 = 0$. The $\cos^2 \beta$ factor in (17c) accounts for the Poynting vector walkoff at angle $\beta$ of the pump field.

An improvement in the coupled equations range of validity and a mathematical convenience occurs if one accounts for the slight crystal dispersion contained in $\epsilon$. The frequency dependence of $\epsilon$ contained in the definition of the linear part of the polarization [15] is written as

$$P_{\text{lin}}(t, z) = \epsilon_0 \int_{-\infty}^{\infty} \chi(\omega) E(\omega, z) e^{i\omega t} d\omega$$  \hspace{1cm} (18)

where the field is represented as

$$E(t, z) = A(t, z) \exp (+i(\omega t - kz)).$$  \hspace{1cm} (19)

The Fourier transform of (19) with a Taylor series expansion of $k$ and $\chi$, about the center frequency $\omega_0$, when substituted into (18) gives to the first order

$$P_{\text{lin}}(t, z) = \epsilon_0 \chi(\omega_0) A(t', z)$$

where

$$t' = t - (n/c) \left[ 1 + (\omega/[2\epsilon]) \frac{d\omega}{d\epsilon} \right] z.$$  \hspace{1cm} (20a)

Previous $\Delta k$ derivations arising from $e^{+i(\omega t - kz)}$ factors remain the same while envelope quantities now are dependent on $t'$ rather than $t$. Thus the phase velocity terms $1/v_p = n/c$ are replaced by the group velocity terms

$$1/v_s = \frac{\partial k}{\partial \omega} = (n/c) \left[ 1 + (\omega/[2\epsilon]) \frac{d\omega}{d\epsilon} \right]$$

in (17). Group velocity considerations are primarily of interest for operation of parametric amplifiers with short pulses in the region of the tens of picoseconds [16]. The group velocities for the coupled waves in the present experiments are equal to within 2 percent so that

$$v_{g_1} = v_{g_2} = v_{g_3} = v_g$$  \hspace{1cm} (21)

is a good approximation for nanosecond pulses.

The change from phase velocity to group velocity in the left-hand side of (17) makes a reduction to ordinary differential equations possible using a transformation indicated by Scott [17]. The transform equations are

$$\begin{align*}
 z &\longrightarrow r = z \\
t &\longrightarrow t = \tau - z/v_g \\
\frac{\partial}{\partial z} &\longrightarrow \frac{\partial}{\partial r} - \frac{1}{v_g} \frac{\partial}{\partial \tau} \\
\frac{\partial}{\partial t} &\longrightarrow \frac{\partial}{\partial \tau}
\end{align*}$$

with a resultant simplification of the coupled equations to

$$\begin{align*}
\frac{\partial E_2}{\partial r} &= -j \frac{\omega_2 d_{\text{eff}}}{n_2 c} E_1^* E_3 e^{-i\Delta kr} \\
\frac{\partial E_3}{\partial r} &= -j \frac{\omega_3 d_{\text{eff}}}{n_3 c \cos^2 \beta} E_1 E_2 e^{i\Delta kr}.
\end{align*}$$

The use of $E_i = \rho_i e^{-i\theta_i}$ with $i = 1, 2,$ and $3$ accomplishes a rectangular to polar dependent variable conversion. The polar form equations that result are

$$\begin{align*}
\frac{\partial \rho_1}{\partial r} &= -\frac{\omega_1 d_{\text{eff}}}{n_1 c} \rho_2 \rho_3 \sin \theta \\
\frac{\partial \rho_2}{\partial r} &= -\frac{\omega_2 d_{\text{eff}}}{n_2 c} \rho_1 \rho_3 \sin \theta \\
\frac{\partial \rho_3}{\partial r} &= +\frac{\omega_3 d_{\text{eff}}}{n_3 c \cos^2 \beta} \rho_1 \rho_2 \sin \theta \\
\frac{\partial \theta}{\partial r} &= \Delta k + \frac{d_{\text{eff}}}{c} \left( \frac{\omega_3}{n_3 \cos^2 \beta} - \frac{\rho_1 \rho_2}{n_2} - \frac{\rho_1 \rho_3}{n_1} \right) \cos \theta
\end{align*}$$

where

$$\theta = \Delta k + \phi_3 - \phi_2 - \phi_1.$$  \hspace{1cm} (24a)

An invariant for the parametric conversion process representing the power flow per unit area parallel to the direction of propagation is

$$W = (e_0 c/2)(n_1 \rho_1^2 + n_2 \rho_2^2 + n_3 \rho_3^2 \cos^2 \beta)$$  \hspace{1cm} (24b)

or

$$W = I_1(0) + I_2(0) + I_3(0) \cos^2 \beta.$$  \hspace{1cm} (24b)

Introducing a set or normalized dependent and independent variables given by

$$\begin{align*}
u_1 &= \left( \frac{e_0 \lambda_1 n_1}{4\pi W} \right)^{1/2} \\
u_2 &= \left( \frac{e_0 \lambda_2 n_2}{4\pi W} \right)^{1/2} \\
u_3 &= \left( \frac{e_0 \lambda_3 n_3 \cos^2 \beta}{4\pi W} \right)^{1/2}
\end{align*}$$

and

$$\frac{4d_{\text{eff}} \pi (\pi W)^{1/2} r}{(e_0 \lambda_1 \lambda_2 \lambda_3 n_1 n_2 n_3 \cos^2 \beta)^{1/2}}$$

where

$$\Delta S = \frac{\Delta kr}{\xi}$$

results in the normalized coupled equations

$$\frac{du_1}{d\xi} = -u_2 u_3 \sin \theta$$  \hspace{1cm} (26a)
\[
\frac{du_2}{d\xi} = -u_1u_3 \sin \theta \tag{26b}
\]
\[
\frac{du_3}{d\xi} = +u_1u_2 \sin \theta \tag{26c}
\]
and
\[
\frac{d\theta}{d\xi} = \Delta S + \left( \frac{u_1u_2}{u_3 - u_1u_3} - \frac{u_2u_3}{u_1 - u_2u_3} \right) \cos \theta. \tag{26d}
\]

The phase equation may also be written as
\[
\frac{d\theta}{d\xi} = \Delta S + \cos \theta \left( \frac{d}{d\xi} \left[ \ln(u_1u_2u_3) \right] \right). \tag{26e}
\]

The phase equation can be integrated with a substitution from (26c) to give
\[
\cos \theta = \left( \Gamma - \frac{1}{2} \Delta S u_3^2 \right)/(u_1u_2u_3) \tag{27}
\]
where
\[
\Gamma = u_1(0)u_2(0)u_3(0) \cos \theta(0) + \frac{1}{2} \Delta S u_3^2(0) \tag{27a}
\]
is a constant of integration.

Energy conservation, power conservation, and momentum conservation for the three-wave interactions are given by the three auxiliary equations
\[
\omega_3 = \omega_1 + \omega_2, \tag{28}
\]
\[
1 = \omega_1 u_1^2 + \omega_2 u_2^2 + \omega_3 u_3^2 \tag{28a}
\]
and
\[
\Delta k = \frac{\xi}{r} \left[ \frac{d\theta}{d\xi} - \left( \frac{u_1u_2}{u_3} - \frac{u_2u_3}{u_1} - \frac{u_3u_1}{u_2} \right) \right] \cos \theta. \tag{28b}
\]

Equation (28) is a normalized version of \( W \). The relative phase between the waves is related to momentum conservation through \( \Delta k \) by (28d). Additional invariants over \( \xi \) are derived by substitution of (28) into (28) giving
\[
m_1 = u_1^2 + u_2^2 \tag{29a}
\]
\[
m_2 = u_2^2 + u_3^2 \tag{29b}
\]
\[
m_3 = u_3^2 - u_2^2 \tag{29c}
\]
which are the Manley-Rowe [18] relations.

The solution of the normalized coupled equations is based on integrating (26c) after substituting the integration constant \( \Gamma \) and the Manley-Rowe relations. The resultant integral is
\[
\xi = \frac{1}{2} \int_{u_3(0)}^{u_3(\xi)} \frac{d(u_3^2)}{u_3^2(m_2 - u_3^2)(m_1 - u_3^2) - (\Gamma - \frac{1}{2} \Delta S u_3^2)^2}^{1/2}. \tag{30}
\]

The cubic polynomial in the denominator radical suggests the use of elliptic integrals as solutions. However, several algebraic manipulations are required to produce a standard elliptic integral form. The denominator polynomial has roots that may be ordered as \( u_3^2 > u_3^2 > u_3^2 > 0 \). Two algebraic substitutions
\[
y^2 = (u_3^2 - u_3^2)(u_3^2 - u_3^2) \tag{31a}
\]
and
\[
\gamma^2 = (u_3^2 - u_3^2)(u_3^2 - u_3^2) \tag{31b}
\]
convert the integral expression to standard elliptic integrals [19]. By partitioning \( \xi \) into \( (\xi + \xi_0) - \xi_0 \), the elliptic integral can be written with zero lower limits as
\[
\xi = \frac{1}{(u_3^2 - u_3^2)} \left( \int_{0}^{\gamma(\xi)} \frac{dy}{[1 - y^2(1 - \gamma^2 y^2)]^{1/2}} \right). \tag{31c}
\]

The inverse operation of the elliptic integral is a Jacobian elliptic function. The above equation rewritten with the Jacobian elliptic functions becomes
\[
y(\xi) = \text{sn} \left[ (u_3^2 - u_3^2)^{1/2}(\xi + \xi_0), \gamma \right] \tag{32a}
\]
\[
y(\xi) = \text{sn} \left[ (u_3^2 - u_3^2)^{1/2}(\xi_0), \gamma \right]. \tag{32b}
\]

The general solution for the normalized \( u \) values is
\[
u_3^2(\xi) = u_3^2 + (u_3^2 - u_3^2)\text{sn}^2 \left[ (u_3^2 - u_3^2)^{1/2}(\xi + \xi_0), \gamma \right] \tag{33a}
\]
\[
u_3^2(\xi) = [m_2 - u_3^2(\xi)] = u_3^2 + u_3^2 - u_3^2(\xi) \tag{33b}
\]
\[
u_3^2(\xi) = [m_1 - u_3^2(\xi)] = u_3^2(0) - u_3^2(\xi) \tag{33c}
\]
where \( \xi_0 = F(\sin^{-1}[(y(0)], \gamma)/(u_3^2 - u_3^2)^{1/2} \) and \( F(\phi, \gamma) \) is a standard elliptic integral.

C. Special Case Solutions

In this section the solutions to the coupled equations given by (33a), (33b), and (33c) are investigated for boundary conditions appropriate to upconversion, sum and second-harmonic generation, mixing and parametric amplification. Appropriate approximations allow the elliptic functions to be reduced to circular and hyperbolic functions in the nonpump depleted regime.

The frequency upconversion process is described by the input conditions
\[
u_3^2(0) = 0 \tag{34a}
\]
\[
u_3^2(0) \ll u_1^2(0) \tag{34b}
\]
causing
\[
u_3^2 = 0 \text{ and } \Gamma = 0 \tag{35}
\]
where \( \omega_3 \) is the sum frequency, \( \omega_1 \) is the frequency of the highest intensity wave, and \( \omega_2 \) is the lowest frequency which is upconverted. The upconversion solutions are
\[
u_3^2(\xi) = u_1^2(0)\text{sn}^2 \left[ u_1(0)(\xi + \xi_0), \gamma \right] \tag{36a}
\]
\[
u_3^2(\xi) = u_1^2(0)(1 - \text{sn}^2 \left[ u_1(0)(\xi + \xi_0), \gamma \right]) \tag{36b}
\]
\[
u_1^2(\xi) = u_1^2(0) - u_1^2(0)\text{sn}^2 \left[ u_1(0)(\xi + \xi_0), \gamma \right]. \tag{36c}
\]
Equations (31) and (34) yield $\gamma^2 = u_2^2(0)/u_1^2(0) \ll 1$ making possible the approximation $\text{sn}(u, \gamma) \approx \sin u$ for $\gamma^2 \ll 1$. Since $y(0) = 0$ from (32) and (34) and $\xi_0 = 0$, inverting the normalized variables given by (25) yields the upconversion solutions

$$I_3(r) \approx (\omega_3/\omega_2) \frac{I_2(0)}{\cos^2 \beta} \sin^2 \left( r/l_u \right)$$  \hspace{1cm} (35a)$$

and

$$I_2(r) \approx I_2(0) \cos^2 \left( r/l_u \right)$$

$$I_1(r) \approx I_1(0)$$  \hspace{1cm} (35c)$$

where

$$1/l_u = u_1(0)\xi_0/r = \frac{\pi d_{\text{eff}}[8I_1(0)]^{1/2}}{(e_0/\lambda_1 \lambda_3 n_1 n_2 n_3 c \cos \beta)^{1/2}}.$$  

For sum generation we assume the input conditions

$$u_1^2(0) = u_2^2(0)$$  \hspace{1cm} (36a)$$

$$u_3^2(0) = 0$$  \hspace{1cm} (36b)$$

resulting in a denominator polynomial inside the radical for (30) of

$$u_3^2(m_1 - u_3^2).$$

The integration simplifies to

$$\xi = \int_{u_1(0)}^{u_3(0)} \frac{du_3}{m_1 - u_3^2} = [1/\xi_0(0)] \tanh^{-1} [u_3(\xi)/u_1(0)]$$

with the result that

$$I_3(r) \approx (\omega_3/\omega_1) \frac{I_1(0)}{\cos^2 \beta} \tanh^2 \left( r/l_u \right)$$  \hspace{1cm} (37a)$$

$$I_2(r) \approx I_2(0) \sech^2 \left( r/l_u \right)$$

$$I_1(r) \approx I_1(0) \sech^2 \left( r/l_u \right)$$  \hspace{1cm} (37c)$$

with

$$I_2(0)/I_1(0) = \omega_2/\omega_1.$$  

Sum generation reduces to second-harmonic generation if the further restrictions $\omega_2 = \omega_1$, $\omega_3 = 2\omega$ and $I_1(0) = I_2(0) = \frac{1}{2} I_3(0)$ are applied yielding

$$I_2(\omega) = \frac{I_2(0)}{\cos^2 \beta} \tanh^2 \left( r/l_{\text{SH}} \right)$$

$$I_1(\omega) = \frac{I_1(0)}{\sech^2 \beta} \left( r/l_{\text{SH}} \right)$$

$$1/l_{\text{SH}} = \frac{\pi d_{\text{eff}}[8I_1(0)]^{1/2}}{(e_0 n_1 n_2 n_3 c \lambda_1 \lambda_2 c)^{1/2}} \cos \beta.$$  

This is the familiar SHG solution which is more directly derived through an energy conservation argument.

**D. Parametric Amplification**

Parametric amplification of energy at $\omega_1$ by a higher intensity at $\omega_3$ is represented by

$$u_3(0) = 0$$

$$u_1^2(0) < u_2^2(0)$$  \hspace{1cm} (39b)$$

with

$$u_{3a}^2 = 0, u_{3b}^2 = m_2 = u_3^2(0), u_{3c}^2 = m_1 = u_3^2(0) + u_1^2(0),$$

and

$$\Gamma = 0.$$  

Since $y(0) = 1$ from (31a), the elliptic integral becomes

$$F[\sin^{-1} y(0), \gamma] = F(\pi/2, \gamma) = K(\gamma)$$

and consequently

$$\xi_0 = K(\gamma)[u_3^2(0) + u_1^2(0)]^{1/2}$$

where $K(\gamma)$ is a complete elliptic integral [19].

The identity $\text{sn}(u + 2K(\gamma), \gamma) = \text{sn}(u, \gamma)$ transforms (33a) to

$$u_3^2(\xi) = u_3^2(0) \text{sn}^2 \left[ u_3(0)(\xi - \xi_0)/\gamma \right], \gamma$$

$$\gamma^2 = u_3^2(0)/[u_3^2(0) + u_1^2(0)].$$

For convenience since $\beta$ is a small angle, we set $\cos^2 \beta = 1$. The parametric amplification solutions which follow from (25), (33), and (40) are

$$I_1(r) = I_1(0) + (\omega_1/\omega_3) I_3(0)(1 - \text{sn}^2 [(r - r_0)/l, \gamma])$$

$$I_2(r) = (\omega_2/\omega_3) I_3(0)(1 - \text{sn}^2 [(r - r_0)/l, \gamma])$$

$$I_3(r) = I_3(0) \text{sn}^2 \left[ (r - r_0)/l, \gamma \right]$$  \hspace{1cm} (41c)$$

where

$$1/l = \mu_3(0) \xi(\gamma/r)$$

$$= \frac{\pi d_{\text{eff}}[8I_3(0)]^{1/2}}{(e_0 n_1 n_2 n_3 c \lambda_1 \lambda_2 c)^{1/2}} \{1 + I_3(0)(\omega_2/[I_3(0)\omega_1])^{1/2}\}$$

and

$$r_0/l = \mu_3(0) \xi_0/\gamma = K(\gamma) = \frac{1}{2} \ln (16 [1 + I_3(0)\omega_1/I_3(0)\omega_3]).$$

where (25d) and the above expression for $\xi_0$ with an expansion of $K(\gamma)$ using logarithm [19] have been used.

For small values of $\xi$ where pump depletion is not important and

$$u_1^2, u_2^2 \ll u_3^2$$

further equation development is useful. A combination of (25) and (40) results in ($\omega_1/\omega_3$)$I_3(0) = I_1(0)\gamma^2/(1 - \gamma^2)$ which modifies (41) to

$$I_1(r) = I_1(0) + \left[ \gamma^2/(1 - \gamma^2) \right] I_1(0)(1 - \text{sn}^2 [(r - r_0)/l, \gamma])$$

$$I_2(r) = (\omega_2/\omega_1) \left[ \gamma^2/(1 - \gamma^2) \right] I_3(0)(1 - \text{sn}^2 [(r - r_0)/l, \gamma])$$

$$I_3(r) = I_3(0) \text{sn}^2 \left[ (r - r_0)/l, \gamma \right].$$

Identities of Jacobian elliptic functions followed by the approximation $\gamma^2 = 1$ permit the transformation

$$\{\gamma^2/(1 - \gamma^2)\} \{1 - \text{sn}^2 [(r - r_0)/l, \gamma]\} = \gamma^2 \text{sn}^2 (r/l) d\text{sn}^2 (r/l) = \sinh^2 (r/l).$$
The results are the familiar relations describing amplification in the parametric approximation given by

\[ I_1(r) = I_1(0) \cosh^2 \left( r/l \right) \]
\[ I_2(r) = I_2(0) (\omega_1/\omega_2) \sinh^2 \left( r/l \right) \]
\[ I_3(r) = I_3(0) \]

This result agrees with the nondepleted pump parametric amplification relations previously derived by Harris [8] and Byer [11].

The parametric amplification case with \( \Delta k \neq 0 \) for the range

\[ \Delta kl < 2 \]

and \( u_1^2, u_2^2 \ll u_3^2 \) is also of interest.

The denominator polynomial of (30) for \( \Delta k \neq 0 \) becomes

\[ u_3^2 (m_2 - u_3^2) (m_1 - u_3^2) - (\Delta S/2)^2 (m_2 - u_3^2)^3 \]

Except for the change from \( u_3^2 = 0 \) to \( u_3^2 = (\Delta S/2)^2 \) the development proceeds in the same manner as the \( \Delta k = 0 \) solution. The \( r/l \) in (44) becomes

\[ r/l = (u_3^2(0)/[\xi/r])^2 - (\Delta k/2)^2 \]

Introducing the parametric gain coefficient \( \Gamma_0 \) defined by

\[ \Gamma_0^2 = u_3^2(0)/[\xi/r] = \frac{\pi^2 d_{\text{eff}}^2 8 I_3(0)}{e_0 \lambda_1 \lambda_2 n_1 n_2 n_3 c} \]

allows the solutions to be written in a previously derived form [10] as

\[ I_1(r) = I_1(0) \cosh^2 \left[ \Gamma_0^2 - (\Delta k/2)^2 \right]^{1/2} r \]
\[ I_2(r) = (\omega_2/\omega_1) I_1(0) \sinh^2 \left[ \Gamma_0^2 - (\Delta k/2)^2 \right]^{1/2} r \]
\[ I_3(r) = I_3(0) \]

### III. COMPUTER METHODS AND EXAMPLE SOLUTIONS

Evaluation of (26) was accomplished by digital calculation. A program was developed to calculate \( I_1, I_2, I_3, \) and \( \theta \) assuming plane waves with user supplied initial values. To initialize the program, values for crystal length, wavelengths, refractive indexes, nonlinear coefficient, and walkoff angle were entered. The finite difference technique used in the computer solution provided a smooth transition between regions of no significant pump depletion and the high conversion efficiency regime. A computer solution based on the evaluation of elliptic integrals for the general analytic solution had difficulties in the region of small pump depletion as noted by Bey and Tang [20].

A computer calculation for a parametric amplifier with only a signal wave input, in addition to the pump, is plotted in Figs. 1(a) and 1(b). Fig. 1(a) represents the relative photon fluxes versus the normalized conversion length parameter \( \xi = r/l \) with the associated relative phase plotted in Fig. 1(b). These plots are solutions of (41). The \( P_i \) values are the \( u_i^2(\xi) \) values normalized by \( u_3^2(0) \).

The relative signal to pump wave intensity ratio is arbitrarily chosen in a range relevant to practical parametric amplifier situations. At the optimal conversion point \( \xi_0 \), two practical examples of the pump intensity help calibrate \( \xi \). The value of \( d_{\text{eff}} \) used for LiNbO\(_3\) is 5.58 \( \times \) 10\(^{-12} \) m/V (\( \lambda_1 = 1550 \) nm, \( \lambda_2 = 3400 \) nm) and for KD\(^{\text{P}}\)P is 5.01 \( \times \) 10\(^{-13} \) m/V (\( \lambda_1 = 580 \) nm, \( \lambda_2 = 913 \) nm). Appendix II discusses a Millers' \( \Delta \) derivation of the \( d_{\text{eff}} \) values needed to calculate \( d_{\text{eff}} \) in (9) and (10).

A computer simulation of parametric amplification for the interesting case where both \( P_1(0) \) and \( P_2(0) \) inputs in addition to \( P_3(0) \) are applied to the parametric medium is plotted in Fig. 2. The input ratios were arbitrarily chosen to be \( P_1(0)/P_2(0) = 1.89 \times 10^{-3} \) and \( P_2(0)/P_3(0) = 1.59 \times 10^{-3} \) which are near typical experimental values. Fig. 2(a) is the magnitude of the relative photon fluxes while Fig. 2(b) plots the relative phase from an arbitrary initial value of \( \pi/4 \). A study of (26d) for various input conditions suggests a first-order approximation of the phase as a linear function of \( \xi \) from the initial value \( \theta(0) \) to \(-\pi/2\). The value of \( \xi_1 \), the approximate point where \( \theta \) first becomes \(-\pi/2\), may be estimated as

\[ \xi_1 = -[\pi/2 + \theta(0)] \frac{d\theta}{d\xi} \bigg|_{\xi=0} \]

After the exponential conversion region the pump is depleted and the phase changes to \( +\pi/2 \) to begin the sum conversion process which regenerates the pump. In Fig. 1(b) the initial phase correction process is immediate at \( \xi = 0 \) since \( P_2(0) = 0 \) in contrast to Fig. 2(b).

A second program was developed to describe the parametric interaction for Gaussian time envelope inputs for the three waves. The inputs include user supplied pulsewidths, relative time offsets and energy fluence values. The assumption of equal group velocities for each field permits the inputs to be divided into time intervals with appropriate average intensity.
values. These intensities become inputs for the first computer program. The sequence of outputs are summed to provide an output energy per unit area \( E(\xi) \). A result for this type of calculation as a function of \( \xi \) is shown in Fig. 3. The peak input intensities for the Gaussian envelope fields are in a ratio \( \omega_3 I_3(0)/\omega_1 I_1(0) = 1.41 \times 10^{-2} \). The result gives conversion efficiencies that are more realistic for actual pulsed parametric amplifier operation. The \( Q_1 \) values are time integrated values of \( u_T(\xi)/u_3(0) \). In Section IV another computer calculation is considered which also takes into account the transverse spatial variation of the input waves.

IV. PARAMETRIC AMPLIFIER EXPERIMENTS

A. Introduction

Parametric amplifier experiments were carried out using KD\(^*\)P and LiNbO\(_3\) crystals which provide parametric amplification over a tuning range that extends from 460-1400 nm and 1400-4000 nm. The KD\(^*\)P crystal was pumped with the third harmonic of a Q-switched Nd:YAG laser while the LiNbO\(_3\) crystal was pumped directly at 1064 nm. The parametric gain, proportional to \( d_{ef}(\lambda_1 \lambda_2 \lambda_3) \) was of the same order for both crystals. The crystals were available in large sizes of approximately 1.5 cm diameter by 5 cm long which greatly facilitated the parametric amplifier measurements.

The parametric gain measurements utilized KD\(^*\)P crystals due to the excellent optical quality of available crystals. The LiNbO\(_3\) OPA measurements were carried out in conjunction with a remote air pollution monitoring program which required high energy tunable radiation in the near infrared region [21], [22]. The LiNbO\(_3\) OPA served as a final conversion stage for a 1064 nm pumped LiNbO\(_3\) parametric oscillator source. The experiment emphasized the overall conversion efficiency to tunable output.

B. KD\(^*\)P OPA Experiments

The KD\(^*\)P OPA measurements were conducted to verify calculated OPA gain values and the predicted angle tuning curve. Fig. 4(a) shows a schematic of the experimental configuration used to measure parametric gain. Fig. 4(b) shows the experimental setup which used a dye laser input to verify the KD\(^*\)P tuning characteristics. In both cases, the laser source consisted of an unstable resonator Nd:YAG oscillator/amplifier system which generated up to 700-mJ 7-11s 1064 pm pulses at ten pulses per second [23].

For the KD\(^*\)P gain measurements, 5 percent of the 1064 nm beam was selected by a beam splitter and transmitted through two polarizers for variable intensity control. The beam was then transmitted through a two to one beam reducing telescope which was carefully adjusted to provide a well collimated idler (\( \omega_2 \)) beam incident on the KD\(^*\)P OPA crystal. The majority of the 1064 nm energy was doubled in a 2.5 cm KD\(^*\)P Type II angle phase matched crystal producing 250 mJ pulses at 532 nm. The remaining 1064 nm energy was summed with the 532 nm output in a 5 cm KD\(^*\)P Type II angle phase-matched crystal to produce 60-mJ 6.5-ns pulses of 355 nm energy. A prism disperser separated the wavelengths. The horizontally polarized 355 nm pumping beam was reduced in diameter and collimated by a 1.7 to 1 telescope. The pump and input idler beams were combined, passed through the KD\(^*\)P Type I angle phase-matched parametric amplifier crystal, and separated with a prism prior to detection. Small uniform intensity regions with an area of 7.85 X
The gains obtained from the measurement of plane wave small area cases are compared with theory in Fig. 5. The dotted line is the intensity gain calculated using the nondepleted pump approximation given by (47). The solid line corresponds to intensity gains calculated with the input idler and pump fluences as input information for the time dependent computer calculations described in Section III. The values of the calculation, including time dependence and pump depletion, are in agreement within the experimental error of the measurements. Pump depletion becomes more significant at higher intensities consistent with the increasing separation between observed results and the nondepleted pump wave approximation.

A slightly modified experimental arrangement shown in Fig. 4(b) was used to perform the KD*P tuning curve verification. The 532 nm energy remaining after the summing crystal was used to side pump a dye laser oscillator [25]. The pulsed dye laser provided an input signal wave source for the parametric amplifier. The dye laser generated 5 ns pulses of 0.2 to 0.5 mJ energy which were tunable from 550 to 680 nm. The idler wave generated in the parametric amplifier tuned from 1000 to 742 nm. The 355 nm pump beam was reduced by a 1.7 to 1 telescope before combining with the dye laser. The spatial overlap of the dye laser Gaussian mode and the unstable resonator pumping beam produced modest signal wave gains of 1.5, quite adequate for tuning curve verification.

Fig. 6 shows a comparison of the calculated and measured phase-matching points for KD*P. The phase-matching curve was determined from a Sellmeier expression [26] for the $n_0$ and $n_e$ indexes of refraction (see Appendix II), combined with the phase-matching condition given by (4). The small walkoff angle correction given by (7) was included.

Computer calculations indicate that a substantial amount of output energy should be available from a 5 cm KD*P parametric amplifier pumped with 355 nm. A 110 mJ pumping beam with an unstable resonator transverse profile and a Gaussian pulsewidth of 6.5 ns was modeled by a set of three concentric rings of graduated intensities. The 2 mJ dye input pulses at 580 nm with a Gaussian pulsewidth of 5 ns used the same ring model. The initial calculations ignored pump beam walkoff. A second calculation reexamined the energy contribution to the output from the most intense dye ring taking into account the walkoff displacement of the pump from...
the signal and idler. Without walkoff, calculations yielded 26.7 mJ signal wave and 15.6 mJ output at an idler wavelength of 913 nm. This is an overall conversion efficiency of 38.5 percent. The central dye ring contributed 17.4 mJ of the signal and 10.3 mJ of the idler output energy. With Poynting vector walkoff included, the central dye input ring contributed 15.1 mJ of signal and 8.9 mJ of idler output energy for an efficiency of 20 percent.

These calculated results show that a carefully designed KD*P parametric amplifier can convert a significant fraction of the input pump energy to tunable signal and idler output energies.

C. LiNbO₃ OPA Experiments

The LiNbO₃ OPA experiments were designed to measure the OPA gain and energy conversion efficiency. Fig. 7(a) and (b) shows a schematic of the LiNbO₃ OPA experiments with a LiNbO₃ OPO and stimulated Raman scattering input sources. The angle tuning curve for 1.064 µm pumped LiNbO₃ extends from 1400-4000 nm as shown in Fig. 8. The design and operational characteristics of the LiNbO₃ OPO are considered in detail in a recent paper [27]. For the present OPA measurements the LiNbO₃ OPO was tuned to an operating wavelength of 1900 nm for comparison with measurements which used the stimulated Raman converter as the input source.

The LiNbO₃ parametric oscillator operated at 56 mJ input 1064 nm energy at two times above threshold. The singly resonant parametric oscillator utilized a double passed pump beam to reduce threshold and improve the output stability. To avoid feedback into the Nd:YAG source, a Faraday rotator isolator was employed prior to the 1.5-1 beam reducing telescope and LiNbO₃ OPO. The generated OPO output was transmitted through a polarizer pair for variable attenuation prior to the LiNbO₃ OPA.

The LiNbO₃ OPA was pumped by 350 mJ 8 ns 1064 nm pulses with the full 6.3 mm diameter near-field unstable resonator beam. The smaller area 0.13 cm² OPO signal beam was incident directly into the OPA to avoid extra optical components. Thus the input beam did not utilize the full 0.30 cm² OPA gain region in this experiment. A dichroic beam splitter was used after the OPA to divert the residual 1064 nm pump energy prior to a power meter which measured the total signal and idler generated output energy.

Fig. 9(a) shows the total OPA output energy available as a function of input signal wave energy for pump characteristic intensities of 52 and 69 MW/cm². Fig. 9 shows OPA gain saturation by plotting the total signal plus idler energy output divided by signal wave energy input versus the signal wave energy input. The 52 MW/cm² pump intensity provides a gain of 55, while a gain of 136 is realized for a 60 µJ input in
both cases. The LiNbO₃ OPO followed by an LiNbO₃ OPA provides the capability of efficiently converting input pump energy to tunable output energy in a manner that is analogous to dye oscillator/amplifier sources.

A 1900 nm signal wave source generated by stimulated Raman scattering in hydrogen gas was also used as an input to the OPA. The experimental arrangement was modified as shown in Fig. 7(b) by substituting a long focal length telescope and the 1 m H₂ cell for the Faraday rotator, beam reducing telescope, and parametric oscillator. In this case, a 6 cm long LiNbO₃ OPA crystal was used. Available optics permitted a better transverse spatial overlap between the input Gaussian signal wave with spot size \( \omega_0 = 0.5 \text{ cm} \) and the unstable resonator pump wave than in the OPO-OPA experiment. The 1 m Raman cell, using fused silica windows, operated at 20 atm H₂ pressure. The threshold for the first vibrational Stokes wave was 18 mJ for a 1064 nm beam focused 50 cm into the cell.

Fig. 9(b) shows an unsaturated gain value of 140 for a 73 MW/cm² pump intensity. This gain value was adjusted to an equivalent gain for a 5.5 cm long OPA crystal used in the OPO experiment. The change in transverse beam overlap significantly improved the total output energy available at higher input energies since more of the available pumping beam area is now utilized.

Table I shows the measured parametric amplifier energy conversion efficiencies from the pump to signal and idler waves for various input pump energies. The measurements were done with \( \lambda_1 = 1900 \text{ nm} \) and a pump beam area of 0.45 cm². The conversion efficiency increases from 10 to 20 percent for pump intensities from 57.5 to 70 MW/cm². The effect of the improved transverse spatial overlap between the parametric

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**TABLE I**

<table>
<thead>
<tr>
<th>Pump Energy (mJ)</th>
<th>Pump Intensity (MW/cm²)</th>
<th>Signal Source</th>
<th>Input Energy (mJ)</th>
<th>Conversion Efficiency (%)</th>
<th>Total Output Energy (mJ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>213</td>
<td>57.5</td>
<td>O.P.O.</td>
<td>0.38</td>
<td>3.3%</td>
<td>7.0</td>
</tr>
<tr>
<td>213</td>
<td>57.5</td>
<td>O.P.O.</td>
<td>0.75</td>
<td>4.7%</td>
<td>10.0</td>
</tr>
<tr>
<td>213</td>
<td>57.5</td>
<td>O.P.O.</td>
<td>1.5</td>
<td>6.6%</td>
<td>14.0</td>
</tr>
<tr>
<td>257</td>
<td>69</td>
<td>O.P.O.</td>
<td>2.0</td>
<td>9.3%</td>
<td>24</td>
</tr>
<tr>
<td>226</td>
<td>73</td>
<td>Raman Cell</td>
<td>3.0</td>
<td>19.5%</td>
<td>44</td>
</tr>
<tr>
<td>226</td>
<td>73</td>
<td>Raman Cell</td>
<td>4.25</td>
<td>22.6%</td>
<td>51</td>
</tr>
</tbody>
</table>

*Input signal area = 0.13 cm².

**Input signal area = 0.82 cm².
The possibility of extracting additional infrared output energy and more conversion efficiency from the pump laser beam was confirmed by an experiment with two sequential parametric amplifier crystals. The experimental configuration was similar to Fig. 7(a) but with the polarizers between the parametric oscillator and amplifier source removed. A 4.1 cm crystal was located after the pump beam and input beam combining beam splitter followed by a second 5.5 cm long LiNbO₃ crystal. The amplified signal plus idler energy was measured following the 1064 nm pump beam diverter. All three waves traveled directly from the first crystal to the second. The signal wavelength was chosen as 1900 nm with the idler at 2420 nm. The 8 ns pump pulses had an energy of 248 mJ/pulse with a pump beam area of 0.45 cm². The measurement results are shown in Table II. The second crystal does provide an increase in conversion efficiency from 4 to 13 percent for the signal wave driven case. In addition, the removal of the polarizer permitted the excitation of the first LiNbO₃ crystal by both the signal and idler waves from the parametric oscillator. A comparison between the two crystal sequence excited by the signal wave only from the parametric oscillator and signal plus idler shows an increase from 32 mJ output to 43 mJ with a conversion efficiency change from 13 to 17.4 percent.

Fig. 10 illustrates the input and output spectra for the Raman cell driven parametric amplifier. A 4.0 mJ input signal wave at 1900 nm was used. The pump energy was 262 mJ with an output signal wave energy of 23 mJ. The output linewidth is predominately controlled by the input bandwidth of 0.4 cm⁻¹ with some broadening due to OPA gain saturation.

V. SUMMARY AND CONCLUSIONS

The theoretical derivations of Section II reviewed and extended the coupled wave equation analysis for parametric amplification. The connection between the solutions including pump depletion and earlier derivations assuming non-depleted pump waves was established. A practical method of generating the Jacobian elliptic pump depleted solutions with a computer program was demonstrated. As an example, the case of a parametric amplifier with ω₁, ω₂, and ω₃ inputs where relative phase is important in the conversion process was considered. A second computer code was written which included a Gaussian pulse time dependence for the parametric amplifier inputs. This model gave more realistic conversion efficiency values. Small-area gain measurements in a KD*P parametric amplifier verified the OPA theoretical calculations. The KD*P angle tuning curve for 355 nm pumping was also carefully measured. The computer calculations were extended to include beam transverse profiles which included a first-order correction for pump beam Poynting vector walkoff.

Significant values of gain in a LiNbO₃ parametric amplifier of greater than 50 for both signal and idler waves from an initial signal wave input were demonstrated. The importance of the analytical derivations were underscored by the saturation behavior of the LiNbO₃ parametric amplifier which generated 50 mJ of output energy for a 4 mJ input energy with a conversion efficiency from the pump of 22.5 percent. Finally, additional gain and conversion efficiency was demonstrated by a two crystal parametric amplifier arrangement. The availability of high-optical quality KD*P and LiNbO₃ nonlinear crystals and the high peak power near diffraction limited Nd:YAG pump source has led to a practical demonstration of optical parametric amplifier capability. The OPA advantages include: a wide tuning range; high gain in the forward direction only, thus avoiding the need for isolation; a relatively narrow bandwidth and narrow angle gain profile which avoids superfluorescence problems; and high conversion efficiencies in saturated gain operation for efficient power amplification. These properties make parametric amplifiers useful for amplification of dye lasers, F-center lasers, parametric oscillators, and tunable outputs generated by mixing or summing processes. The advantages of parametric amplifiers...
which are well known in the microwave region should apply equally well in the optical region.

**APPENDIX I**

**CRYSTAL ACCEPTANCE ANGLE AND BANDWIDTH**

The phase velocity mismatch factor is defined by

\[ \Delta k = \frac{\partial n_3(\theta)}{\partial \theta} \Delta \theta + g\theta^2 - b\Delta \omega_1, \]  
(A-1)

where

\[ \frac{\partial n_3(\theta)}{\partial \theta} = \frac{n_\omega n_o (n_\omega^2 - n_o^2) \sin \theta_m \cos \theta_m}{(n_\omega^2 \sin^2 \theta_m + n_o^2 \cos^2 \theta_m)^{3/2}}, \]

\[ g = k_1 k_3 / 2k_2, \]

and

\[ b = \frac{\partial k_1}{\partial \omega_1} - \frac{\partial k_2}{\partial \omega_2}. \]

The geometry of Fig. 11 illustrates the momentum vectors and associated angles. Assuming small angles for \( \phi \) and \( \psi \) and taking into account various contributions to \( \Delta k \) by Taylor's series expansions, the momentum mismatch expression becomes

\[ \Delta k = (\omega_3/c) \frac{\partial n_3(\theta)}{\partial \theta} \delta_\theta + g\theta^2 - b\Delta \omega_1, \]  
(A-2)

The first term represents an \( \omega_3 \) beam acceptance angle in the horizontal plane with the \( \omega_1 \) input beam at angle \( \theta_m \) from the crystal optic axis. The second term is either the \( \omega_3 \) beam acceptance angle in the vertical plane with fixed \( \omega_1 \) or the \( \omega_1 \) acceptance angle in any plane with \( \omega_3 \) fixed. The final term relates \( \Delta k \) to broad linewidth \( \omega_1 \) inputs [28], [29].

**APPENDIX II**

**NONLINEAR COEFFICIENT VALUES BY MILLERS' A SCALING**

The value of \( d(-\omega_3, \omega_0, \omega_o)_{ijk} \) or in condensed notation \( d_{ij} \) (see BK) is estimated by utilizing Miller's \( \Delta \) [30]. The nonlinear tensor element is related to Miller's \( \Delta \) and linear susceptibilities by [31]

\[ d_{ijk} = e_0 \Delta_{ij} \chi_{ii}(\omega_1) \chi_{ij}(\omega_2) \chi_{kk}(\omega_3) \]  
(A-2)

where

\[ \chi_{ii} = (n_\omega^2 - 1) \text{ at } \omega_1; \]
\[ \chi_{ij} = (n_\omega^2 - 1) \text{ at } \omega_2; \]
\[ \chi_{kk} = (n_\omega^2 - 1) \text{ at } \omega_3; \]

and

\[ \Delta_{ij} = 1.13 \times 10^{-2} \text{ for LiNbO}_3 \]  
\[ \Delta_{ijk} = 5.99 \times 10^{-3} \text{ for LiNbO}_3 \]  
\[ \Delta_{ik} = 4.04 \times 10^{-2} \text{ for KD*P} \]

from Choy and Byer [30].

The indexes of refraction for \( \text{LiNbO}_3 \) are available from previous measurements [32], while the index values for \( \text{KD*P} \) have been curve fit to the following Sellmeier equations [26] at \( T = 25^\circ C \) for \( \lambda \) in \( \mu m \).

\[ n_3^2(\text{KD*P}) = 1.012233 + \frac{1.23137}{1.00 - 0.83818 \times 10^{-2}} \]  
\[ \frac{1.00 - 0.2771624}{\lambda^2} \]

\[ n_2^2(\text{KD*P}) = 0.933294 + \frac{1.193722}{1.00 - 0.7481954 \times 10^{-2}} \]  
\[ \frac{1.00 - 0.9423675}{\lambda^2} \]

**ACKNOWLEDGMENT**

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**REFERENCES**

Abstract—Using experimentally determined oscillator strengths and photoionization cross-sectional data, we compute the dispersion characteristics of Ar, Kr, and Xe up to their first ionization levels and determine the spectral regions in the VUV where these gases exhibit negative dispersion and can so be efficiently used for frequency tripling. We then investigate the bandwidths over which efficient tripling can be achieved in phase-matched gas mixtures. The bandwidth is limited by the rapidly varying dispersion in the vicinity of resonance transitions in the gases. In particular, we look at the case of frequency tripling 3647 Å radiation to 1215.7 Å (hydrogen Lyman-α) and show, that for fundamental wavelength bandwidths as narrow as 1 Å, the rapid change in refractive index with wavelength can preclude phase matching over the entire bandwidth of the radiation.

INTRODUCTION

In recent years generation of tunable coherent radiation in the vacuum ultraviolet has been achieved in several systems of vapors and gases by frequency tripling. These systems, like their crystal counterparts, require careful attention to phase matching between the driving and generated beams. Because efficient generation usually demands the use of tight focusing in the center of long cells, it is necessary to use gases which are negatively dispersive between the fundamental and harmonic wavelengths to achieve this phase matching. This negative dispersion is achieved by using the anomalous dispersion.

Third-Harmonic Generation in Argon, Krypton, and Xenon: Bandwidth Limitations in the Vicinity of Lyman-α

RITA MAHON, THOMAS J. McILRATH, VALERIE P. MYERSCOUGH, AND DAVID W. KOOPMAN


