Parametric Amplification in Spatially Extended Media and Application to the Design of Tuneable Oscillators at Optical Frequencies

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A theory of traveling wave and backward wave variable-parameter amplification appropriate to the amplification of a light beam is developed. It is an extension of the theory of Tien and Suhl for one-dimensional propagation to the case in which the pump wave, signal wave, and idler waves have different directions of propagation. The theory is then applied to the design of a tuneable oscillator at optical wavelengths. The device is tuned by changing the orientation of a parallel mirror system. It appears that currently available pulsed laser powers are sufficient to drive such devices and that a continuous tuning range over a three-to-one interval in frequency is possible.

I. INTRODUCTION

THE existence of mixing of light beams in nonlinear dielectric media has now been demonstrated by numerous workers. Both the appearance of second harmonic in a single laser beam[1] and the appearance of a sum frequency for overlapping beams have been demonstrated.[2] Furthermore, the relevance of electromagnetic momentum conservation for the realization of large coherence volumes has been pointed out and the importance of the effect demonstrated experimentally.[3,4] An important application of these phenomena would appear to be traveling wave and backward wave variable-parameter amplification. As we shall show in the next two sections, it appears that this process can be used in the design of tunable oscillators at optical frequencies, and that with currently available laser power levels and dielectric media, quite wide range tunability should be achievable.

II. TRAVELING WAVE AND BACKWARD WAVE VARIABLE-PARAMETER AMPLIFICATION

In this section we develop variable-parameter amplification theory in a form suited to the problem at hand. What is required is an extension of the theory of Tien and Suhl[5] to more than one dimension. To avoid uninteresting complications we shall consider the two-dimensional isotropic case, with polarizations selected in the simplest way.

We consider first an infinite homogeneous dielectric medium whose dielectric constant is modulated by a wave of the form $\cos(\omega_p t-\mathbf{q}_p\cdot\mathbf{r})$. In accordance with the usual terminology, ω_p will be referred to as the pump frequency and $\mathbf{q}_p$ as the pump wave vector. For reasons which will become clear in the next section, we assume no special relation between ω_p^2 and q_p^2. Maxwell's equations then take their usual form:

$$\begin{aligned}\nabla\times\mathbf{E}&=-(1/c)\partial\mathbf{B}/\partial t,\\ \nabla\times\mathbf{B}&=(1/c)\partial\mathbf{D}/\partial t.\end{aligned}\tag{1}$$

The relation between $\mathbf{E}$ and $\mathbf{D}$, however, we write in the form

$$\begin{aligned}\mathbf{D}(\mathbf{r},t)&=\int g(t-t')\mathbf{E}(\mathbf{r},t')dt'+2\epsilon\cos(\omega_p t-\mathbf{q}_p\cdot\mathbf{r})\mathbf{E}(\mathbf{r},t),\\ g(t-t')&=\frac{1}{2\pi}\int K(\omega)e^{i\omega(t-t')}d\omega,\quad K(\omega)=K^*(-\omega).\end{aligned}\tag{2}$$

The first term is the unmodulated part of the dielectric constant expressed in a form which takes dispersion into account. The second term is just the modulated dielectric constant.[5a] We assume that ϵ is small compared to $K(\omega)$.

Equations (1) and (2) have solutions of the form

$$\mathbf{E}=\left(\operatorname{Re}\sum_{n=-\infty}^{\infty}E_n e^{+i(\omega_p t-\mathbf{q}_p\cdot\mathbf{r})n}e^{i(\omega_1 t-\mathbf{q}_1\cdot\mathbf{r})}\right)\hat{e}.\tag{3}$$

In (3) we regard ω_1 as the signal frequency and choose it to be real and less than ω_p. If E_0 is regarded as a free parameter, the remaining complex coefficients are determined by (1) and (2). Since we have assumed ϵ small, only those terms for which

$$(n\mathbf{q}_p+\mathbf{q}_1)^2-[(n\omega_p+\omega_1)^2/c^2]K(n\omega_p+\omega_1)\approx 0\tag{4}$$

will be important. Equations (1) and (2) impose certain restrictions on $\mathbf{q}_1$ (it may even be complex) which will always lead to (4) being satisfied for at least one term. Under certain conditions, to be discussed in more detail later, it may hold for two terms, but will except in some rare circumstances, which are ignored, not hold for more than two terms. The interesting case is, of course, the well-known one for which (4) holds for the signal

[1] P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, Phys. Rev. Letters **7**, 118 (1961).
[2] M. Bass, P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, Phys. Rev. Letters **8**, 18 (1962).
[3] J. A. Giordmaine, Phys. Rev. Letters **8**, 19 (1962).
[4] P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage, Phys. Rev. Letters **8**, 21 (1962).
[5] P. K. Tien and H. Suhl, Proc. Inst. Radio Engrs. **46**, 700 (1958).
[5a] We have exhibited the dispersion of the unmodulated dielectric constant explicitly because of its significance in (4). The modulated part of the dielectric constant is, of course, also frequency dependent, and for quantitative application of the theory its dispersion should also be taken into account.

frequency ω_1, and the so-called idler frequency $\omega_p-\omega_1$, which we shall denote by ω_2. Hence, an appropriate approximate solution will take the form [changing notation slightly from (3)]

$$\mathbf{E}=\mathrm{Re}(E_1 e^{i(\omega_1 t-\mathbf{q}_1\cdot\mathbf{r})}+E_2 e^{i(\omega_2 t-\mathbf{q}_2^*\cdot\mathbf{r})})\hat{e}, \quad (5)$$

with $\omega_1+\omega_2=\omega_p$ and $\mathbf{q}_1+\mathbf{q}_2=\mathbf{q}_p$. Substitution of (5) into (1) and (2) then yields

$$\begin{aligned} &E_1 q_1^2/k_1=k_1K_1E_1+\epsilon k_1 E_2^*, \quad k_i=\omega_i/c;\ K_i=K(\omega_i),\\ &E_2^* q_2^2/k_2=k_2K_2^*E_2^*+\epsilon k_2 E_1, \end{aligned} \quad (6)$$

which has nonzero solutions only if

$$(q_1^2-k_1^2K_1)(q_2^2-k_2^2K_2^*)-\epsilon^2k_1^2k_2^2=0. \quad (7)$$

In accordance with a previous comment we note that (7) implies that at least one of the factors $(q_i^2-k_i^2K_i)$ be small. Amplification occurs, however, only when both of the factors are small. Equation (7) has, of course, many solutions, the $\mathbf{q}_i$ which actually occur in a given situation being determined by boundary conditions. To illustrate what is involved we consider a wave of frequency ω_1, wave vector $\mathbf{q}_{10}$, with $q_{10}^2=k_1^2K_1$ propagating in unmodulated dielectric impinging upon a plane interface between unmodulated and modulated dielectric. This will, in general, generate two wave pairs in the modulated region both with frequencies ω_1,ω_2 and wave vectors $\mathbf{q}_1,\mathbf{q}_2=\mathbf{q}_p-\mathbf{q}_1$ and $\mathbf{q}_1',\mathbf{q}_2'=\mathbf{q}_p-\mathbf{q}_1'$, respectively. In addition, reflected waves of frequency ω_1,ω_2 and wave vectors $\mathbf{q}_{10}',\mathbf{q}_{20}'$ satisfying $(q_{20}'^2-k_2^2K_2^*)=0$ (note: $\mathbf{q}_{20}'$ need not equal $\mathbf{q}_p-\mathbf{q}_{10}'$), will appear in the unmodulated region. The various vectors are determined by the boundary conditions and Eq. (7) in the following way. In order that the continuity conditions on $\mathbf{E}$ and $\mathbf{B}$ be satisfiable at all points on the interface it is necessary that

$$\begin{aligned} &\hat{n}\times\mathbf{q}_{10}=\hat{n}\times\mathbf{q}_1=\hat{n}\times\mathbf{q}_1'=\hat{n}\times\mathbf{q}_{10}',\\ &\hat{n}\times\mathbf{q}_2=\hat{n}\times\mathbf{q}_2'=\hat{n}\times\mathbf{q}_{20}', \end{aligned} \quad (8)$$

where $\hat{n}$ is the interface normal, directed into the modulated region. Hence, $\mathbf{q}_1,\mathbf{q}_2$ (and $\mathbf{q}_1',\mathbf{q}_2'$) must be expressible in the form

$$\begin{aligned} &\mathbf{q}_1=\mathbf{q}_{10}+\gamma\hat{n}, \quad \mathbf{q}_{20}=\mathbf{q}_p-\mathbf{q}_{10},\\ &\mathbf{q}_2=\mathbf{q}_{20}-\gamma\hat{n}. \end{aligned} \quad (9)$$

We assume both γ and $q_{20}^2-k_2^2K_2^*$ small and apply Eq. (7), obtaining

$$\gamma=\frac{q_{20}^2-k_2^2K_2^*}{4\mathbf{q}_{20}\cdot\hat{n}} \pm\left[\left(\frac{q_{20}^2-k_2^2K_2^*}{4\mathbf{q}_{20}\cdot\hat{n}}\right)^2-\frac{\epsilon^2k_1^2k_2^2}{4\mathbf{q}_{10}\cdot\hat{n}\ \mathbf{q}_{20}\cdot\hat{n}}\right]^{1/2}. \quad (10)$$

The unprimed and primed wave vectors correspond, respectively, to the use of the upper and lower signs in (9) and (10). $\mathbf{q}_{10}',\mathbf{q}_{20}'$ are, of course, uniquely determined by the requirements

$$\begin{aligned} &\mathbf{q}_{i0}'\times\hat{n}=\mathbf{q}_{i0}\times\hat{n}, \quad q_{10}'^2-k_1^2K_1=0,\\ &\mathbf{q}_{i0}'\cdot\hat{n}<0, \quad q_{20}'^2-k_2^2K_2^*=0. \end{aligned} \quad (11)$$

For the loss free case, (i.e., K_i real), there will typically exist a frequency range for which the relation $q_{20}^2-k_2^2K_2=0$ can be satisfied exactly for an appropriately selected angle between $\mathbf{q}_{10}$ and $\mathbf{q}_p$. In this case (10) takes the form

$$\gamma=\pm\tfrac{1}{2}i\frac{\epsilon k_1k_2}{(\mathbf{q}_{10}\cdot\hat{n}\ \mathbf{q}_{20}\cdot\hat{n})^{1/2}}. \quad (12)$$

For the case $\mathbf{q}_{20}\cdot\hat{n}>0$, one notes the exponential gain characteristic of traveling wave amplifiers ($\hat{q}_{10}\cdot\hat{n}>0$ always, of course, by definition of $\hat{n}$). It is then reasonably apparent that $(q_{20}^2-k_2^2K_2)/4\mathbf{q}_{20}\cdot\hat{n}$ plays the role of determining an effective bandwidth in both frequency and angle, since, for large enough values of this quantity, γ becomes real. On the other hand, for $\mathbf{q}_{20}\cdot\hat{n}<0$, γ is always real. Nevertheless, amplification is still possible, the behavior now being characteristic of a backward wave amplifier. To fully exhibit these features it is necessary to solve a complete amplifier boundary value problem. That is, one introduces a second interface a distance L from the first one, separating the modulated region from a second unmodulated region. In this second region one again assumes waves of frequency ω_1,ω_2 with wave vectors $\mathbf{q}_{10}$, $\mathbf{q}_{20}''$, $\mathbf{q}_{20}''$ differing from $\mathbf{q}_{20}'$ only in the sign of its normal component. One must also assume additional reflected waves in the modulated region [satisfying Eq. (7) with only one of the factors $(q_i^2-k_i^2K_i)$ small]. Continuity conditions on $\mathbf{E}$ and $\mathbf{B}$ at both interfaces and both frequencies then determine all of the relative amplitudes.

The problem outlined above can be greatly simplified by recognizing that the reflected waves in the modulated region and the reflected wave with wave vector $\mathbf{q}_{01}'$ are always small. In the traveling wave case the wave with wave vector $\mathbf{q}_{02}'$ is also small, while in the backward wave case, that with $\mathbf{q}_{02}''$ is small. The problem is simplified by neglecting all of the small amplitudes and applying continuity conditions to the electric fields only. (We are assuming from this point on that the electric polarization vector $\hat{e}$ has been chosen parallel to the interfaces.) The tangential components of the magnetic field will then contain discontinuities of order ϵ. The removal of these discontinuities would require the inclusion of the neglected waves which are also of order ϵ. Such inclusion would also lead to order ϵ corrections to the approximately determined amplitudes. Since the waves to be neglected with $\mathbf{q}_{20}\cdot\hat{n}$ positive are not the same as those to be neglected with $\mathbf{q}_{20}\cdot\hat{n}$ negative, the question of the transition from one sign to the other arises. More detailed examination shows that the proposed approximation depends upon the assumption: $\gamma/\mathbf{q}_{20}\cdot\hat{n}\ll 1$. Hence, within a narrow region about $\mathbf{q}_{20}\cdot\hat{n}=0$ the approximation is invalid.

For the traveling wave case we have the conditions

$$\begin{aligned} E_1+E_1'&=E_i,\\ E_2{}^*+E_2'{}^*&=0,\\ E_1e^{\Gamma L}+E_1'e^{-\Gamma L}&=E_0, \end{aligned} \tag{13}$$

where E_i is the incident amplitude, E_0 the output amplitude, and

$$\Gamma=(\eta^2-\delta^2)^{1/2},$$

with

$$\eta=\epsilon k_1k_2/2|\mathbf{q}_{10}\cdot\hat{n}\mathbf{q}_{20}\cdot\hat{n}|^{1/2},\quad \delta=q_{20}{}^2-k_2{}^2K_2{}^*/4\mathbf{q}_{20}\cdot\hat{n}. \tag{14}$$

Defining the power gain by $G=|E_0/E_i|^2$, one finds easily that

$$G=1+\frac{\eta^2}{\eta^2-\delta^2}\sinh^2[(\eta^2-\delta^2)^{1/2}L]. \tag{15}$$

For the backward wave case we have the conditions

$$\begin{aligned} E_1+E_1'&=E_i,\\ E_2{}^*e^{-i\Gamma' L}+E_2'{}^*e^{+i\Gamma' L}&=0,\\ E_1e^{-i\Gamma' L}+E_1'e^{i\Gamma' L}&=E_0, \end{aligned} \tag{16}$$

with

$$\Gamma'=(\eta^2+\delta^2)^{1/2},$$

from which we compute

$$G=1\bigg/\left(1-\frac{\eta^2}{\eta^2+\delta^2}\sin^2[(\eta^2+\delta^2)^{1/2}L]\right). \tag{17}$$

In the traveling wave case G peaks at $\delta=0$; for the backward wave case this is also true provided that $\eta L\leq\pi/2$. The peak gain is evidently controlled by

$$\eta L=\epsilon k_1k_2L/2|\mathbf{q}_{10}\cdot\hat{n}\ \mathbf{q}_{20}\cdot\hat{n}|^{1/2}. \tag{18}$$

The angular factors in the denominator require some comment. This formula suggests that one can reduce modulation or length requirements simply by arranging for small values of either $\mathbf{q}_{10}\cdot\hat{n}$, $\mathbf{q}_{20}\cdot\hat{n}$, or both. In order to better understand the role played by these directional factors it is necessary to consider an incident beam of finite width. We continue to treat the modulated region as an infinite plane slab but it is clear that it need, in fact, be present only in the region traversed by the beam. In order for a beam to be amplified in accordance with the theory given above it is necessary that it be broader than some minimum value. The point is that a beam of finite width can be thought of as a superposition of infinite beams incident over a narrow range of angles centered about the beam angle. The wider the beam the narrower the angular spread. If the extreme components in the angular distribution are amplified less than the central components, the beam will, of course, broaden as it is amplified. For a given gain and amplifier design there is a minimum beam width W which must be used, if efficient amplification without excessive broadening is to be achieved. One can readily show that for an amplifier with a specified gain and specified relation among the $\mathbf{q}_p$, $\mathbf{q}_{i0}$ and ω_i, the product $WL'\epsilon^2$, where $L'=L/\hat{q}_{10}\cdot\hat{n}$, is the beam length in the modulated region, has a fixed value, and is, in particular, independent of the direction denoted by $\hat{n}$. On the other hand, W/L', which might be thought of as defining an optimum shape for the interaction region, is independent of ϵ and proportional to $|\hat{q}_{10}\cdot\hat{n}/\hat{q}_{20}\cdot\hat{n}|$.

It is interesting also to consider solutions of the form (5) with the $\mathbf{q}_i$ real and in which the direction of $\mathbf{q}_1$ and $\mathbf{q}_2$ are fixed, say, by a pair of Fabry-Perot mirror systems. Under these conditions $\mathbf{q}_1$ and $\mathbf{q}_2$ are both completely determined by the additional requirement $\mathbf{q}_1+\mathbf{q}_2=\mathbf{q}_p$. In this case the interesting solutions will have complex ω so that ω_2 in (5) should be replaced by $\omega_2{}^*$. Equations (6) and (7) then hold unchanged and (7) is an equation determining ω_1 and $\omega_2=\omega-\omega_1$. On the assumption that the $\mathbf{q}_i$ which have been determined satisfy the restriction $\Delta\omega\ll\omega$, where

$$\Delta\omega=\omega_p-(q_1c/K_1{}^{1/2})-(q_2c/K_2{}^{1/2}), \tag{19}$$

one easily determines

$$\begin{aligned} \omega_1&=q_1/K_1{}^{1/2}+\tfrac{1}{2}\Delta\omega\pm\tfrac{1}{2}i(\epsilon^2\omega_1\omega_2/K_1K_2-\Delta\omega^2)^{1/2},\\ \omega_2{}^*&=q_2/K_2{}^{1/2}+\tfrac{1}{2}\Delta\omega\pm\tfrac{1}{2}i(\epsilon^2\omega_1\omega_2/K_1K_2-\Delta\omega^2)^{1/2}, \end{aligned} \tag{20}$$

corresponding to an oscillation frequency and rate of buildup. Oscillation threshold conditions can easily be deduced by relating the buildup rate to a rate of energy gain and equating it to the reflection loss rate.

III. TUNABLE OPTICAL FREQUENCY OSCILLATORS

Apart from the evident application to amplification, one of the most interesting applications of the process described in the preceding section would appear to be to the design of tunable oscillators operating in the infrared and optical regions.

Let us assume that one has succeeded in modulating a dielectric constant with frequency ω_p and wave vector $\mathbf{q}_p$ or, better, with wave vector $-\mathbf{q}_p$ as well. Suppose further that an interesting frequency range for ω_1 exists such that $|\mathbf{q}_{10}|+|\mathbf{q}_{20}|>|\mathbf{q}_p|$. Then over this range there will exist a unique relation between angle and frequency satisfying the relation $\mathbf{q}_{10}+\mathbf{q}_{20}=\mathbf{q}_p$. By providing a Fabry-Perot mirror system oriented so as to provide a resonant circuit for a particular direction $\mathbf{q}_{10}$, one can make oscillation at the frequencies ω_1 and ω_2 take place. It is only necessary that the beam amplification on each pass through the amplifier exceed the reflection and transmission losses. If both $\mathbf{q}_p$ and $-\mathbf{q}_p$ are present in the dielectric modulation, then amplification takes place for both directions of signal beam traversal. Since the magnitudes of ω_1 and ω_2 depend upon the orientation of the mirror system, the device can be tuned by varying the orientation.

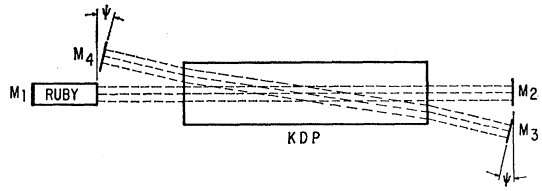

FIG. 1. Schematic arrangement for tunable optical oscillator. The M_i are mirror reflecting surfaces. The optic axis of the potassium dihydrogen phosphate (KDP) is in the plane of the paper and perpendicular to the direction of the ruby laser beam. The [110] crystal axis is perpendicular to the plane of the paper. The laser beam is the extraordinary way. The device is tuned by varying the mirror angle, ψ.

We now briefly discuss the problem of producing the required dielectric-constant modulation. Basically, the modulation is accomplished by passing intense laser beams through the medium. The fact that observations of optical harmonic production have up to now been confined to crystals lacking an inversion point calls attention to the fact that such materials are especially well suited to such applications. Since the dielectric constant of such materials contains terms linear in the electric field, it is reasonable to suppose that a given small dielectric constant modulation can be achieved at lower power levels in such materials than in ordinary materials, for which the dependence is quadratic. The triangle inequality $q_p<q_1+q_2$ then typically requires that the medium be doubly refracting, the faster wave being used for the pump, the slower wave for q_1,q_2. For potassium dihydrogen phosphate (KDP), the material which has been used in this context for second harmonic production,[3,4] the ordinary and extraordinary indices differ by about 2.5% so that the triangle inequality is only weakly satisfied. Normal dispersion will further weaken it. This has the consequence that the angles between vectors are very small except for the case of large inequalities between ω_1 and ω_2. In this latter case one may have difficulty with infrared absorption of the smaller component.[6] For $\omega_1=\omega_2$ the angle required between the pump wave and, say, the direction for ω_1 is of the order of 10°. As the angle is increased, ω_1 will then decrease. The pump power requirements tend to increase as ω_1 and ω_2 become unequal due to the $(\omega_1\omega_2)^{1/2}$ factor in Γ. Furthermore, as ω_1 is decreased, infrared absorption will eventually make further increase in pump power necessary. Nevertheless, the arrangement indicated in Fig. 1 appears to be a satisfactory one. A rectangular prism of KDP with optically flat ends could be incorporated into a ruby laser circuit. The angle to be regenerated is then selected by varying the mirror angles. The minimum angle at which the beam need leave the end of the crystal is of the order of 15° so that interference between the various components appears to be easily avoidable. A tuning range from $(1/4)\omega_p$ to $(3/4)\omega_p$ appears to be quite feasible. An increase in the minimum angle required and a less rapid variation of frequency with angle could be achieved by the use of piezoelectric materials more birefringent than KDP.

In the theory developed in the preceding sections the assumption of optical isotropy was made. In order to discuss the general case of an anisotropic medium with anisotropic modulation some straightforward modifications would be required. For the most useful applications, however, the theory can be applied unchanged. For the arrangements like those described in Fig. 1 it is most convenient to have the laser beam polarized[7] in the plane of the wave vectors $\mathbf{q}_p,\mathbf{q}_1$, and $\mathbf{q}_2$. The crystal should be oriented with its principal axis of least dielectric constant along the electric vector of the pump wave. Then for the case in which $\mathbf{q}_1,\mathbf{q}_2$ are polarized perpendicular to the wave vector plane (case I), the theory of Sec. II applies unchanged. The triangle inequality can generally not be satisfied for $\mathbf{q}_1,\mathbf{q}_2$ both polarized in the wave vector plane. For sufficiently anisotropic materials, however, it may be satisfied for one in the wave vector plane and the other perpendicular to it (case II), in which case two (possibly three) frequencies [with two (three) distinct idler directions] would be possible for a given mirror position. It is possible to favor one case over the other by properly choosing the remaining degree of freedom in the crystal orientation. It is also possible to choose one by inserting a polarizer in the signal circuit or by adding a mirror system to the idler circuit to empahsize one of the idler directions.

It is simplest to make general statements about the case of negative uniaxial crystals (such as KDP). In this case the optic is fixed along the laser beam polarization direction and $\mathbf{q}_1$ and $\mathbf{q}_2$ are then independent of the remaining degree of freedom (the orientation about the optic axis). It is always possible to choose this orientation to optimize case I, which leads to case II being completely forbidden. In some cases crystal symmetry[8] (in spite of the absence of an inversion point) forbids both cases (classes 12, 18, 19, 22, 24). For class 11 (KDP) it determines the orientation uniquely, while for class 9 the optimum orientation must be determined experimentally and may be somewhat frequency dependent. For the remaining classes all orientations are optimum.

[6] With reference to infrared absorption it may be worth noting that the effect of a small but not negligible absorption upon the gain is considerably less if it occurs in the idler circuit rather than in the signal circuit.

[7] We use the convention in which the polarization direction is that of the electric field.

[8] The Von Groth class numbering is used. The remarks about limitations imposed by crystal symmetry are all easily deducible from the table of third rank tensors for the crystal classes, to be found, for example, in W. P. Mason, *Piezoelectric Crystals and Their Applications in Ultrasonics* (D. Van Nostrand Company, Princeton, New Jersey, 1950), pp. 41–44. They hold to the extent that the dipole approximation is valid and to the extent that dispersion in the modulation coefficients can be neglected in the frequency range between ω_1 and ω_2.

For the positive uniaxial crystals the orientation which maximizes the triangle inequality (optic axis perpendicular to the wave vector plane) forbids case I. This means that the triangle equality must be weakened either by employing case II (with two frequencies often possible) or changing the orientation of the optic axis leading to case I as well as case II being allowed with the possibility of three frequencies. It appears therefore that positive uniaxial crystals are less suitable for this application than negative ones.

Biaxial crystals are also applicable, of course, although optimum orientation would typically have to be determined experimentally. It might be remarked that there appear to be many materials which could be regarded as candidates for this application, some of them considerably more anisotropic than KDP.

We turn now to the question of pump power requirements. Reference to Eq. (14) suggests that oscillation should be achievable with ϵ/K in the range 10^{-5} to 10^{-6}, assuming linear dimensions of the order of a few cm. Such modulations can probably be achieved with field strengths[1,8a] of the order of 10^{-5} to $10^{-6}\ e/a_0^2$ or power flux of the order of 10^{+5} to 10^7 watts/cm^2. It should be noted that there is no advantage, as far as oscillation conditions are concerned, in large beam height. Beam heights of the order of a fraction of a millimeter would not lead to excessive diffraction spreading and would reduce the total power requirement. Incorporation of the crystal in the laser circuit as suggested in the previous paragraph also decreases the required power level of the laser. The power level required can be further reduced by improving the reflection efficiency (especially by making some provision to avoid reflection loss at the crystal surfaces) and by adding a pair of mirrors for the idler circuit as well. The last two items would be especially effective in conjunction, and while they might complicate the mechanical arrangements, could lead to substantial decrease in power requirements.

There are, of course, other methods for achieving the required dielectric constant modulation. One such method, which appears to offer considerable flexibility, is based upon the use of two pump beams at frequency ω_{p_1} and ω_{p_2}, in a medium whose dielectric constant contains only terms even in the field strength. The dielectric modulation frequency is then $\omega_p=\omega_{p_1}+\omega_{p_2}$. The magnitude of the pump wave vector $q_p=|\mathbf{q}_{p_1}+\mathbf{q}_{p_2}|$ may have any convenient value less than $q_{p_1}+q_{p_2}$ simply by choosing the relative directions appropriately. Apart from the evident flexibility, one can obtain frequencies in excess of the pump frequency. Such frequencies can, of course, also be obtained tunably by the coherent mixing of the output of lower frequency tunable oscillators. This particular method suffers from the limitation of requiring, on current estimates of nonlinear terms, excessive pump powers. While the required powers will probably be achieved eventually they may very well have destructive effects upon the material. On the other hand, the quadratic effects have not yet been observed and may not be so small as estimated. Furthermore, there is the possibility in suitably selected material, of resonant enhancement at the frequency ω_p, which would not affect the propagation at the pumping beam and signal frequencies.

In this connection one might suggest that the study of three-beam interaction in such material could yield useful information about the size of the quadratic terms. Three beams at frequencies $\omega_1,\omega_2,\omega_3$ with wave vectors $\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}_3$ can interact to produce an output at $\omega_4=\omega_1+\omega_2-\omega_3$ and $\mathbf{q}_4=\mathbf{q}_1+\mathbf{q}_2-\mathbf{q}_3$ provided $q_4^2=k_4^2K_4$. The conditions are identical with those involved in amplification, regarding ω_1 and ω_2 as the pumps, ω_3 as the signal, ω_4 as the idler, the only difference being that production of the idler wave rather than gain of the signal is the effect of interest.[9]

[8a] *Note added in proof.* More recent estimates, based on measurements, reported by R. W. Terhune, P. D. Maker, and C. M. Savage, Phys. Rev. Letters **8**, 404 (1962), indicate that the required field strength may be more than a factor 10^3 greater than that suggested above.

[9] The author has estimated the size of this effect for the quantum electrodynamical nonlinearities of the vacuum, based on the Lagrangian of Euler and Heisenberg. The method offers several advantages over others one might consider. The photons to be detected have a frequency different from those in the sources, and are produced in a well collimated beam. Furthermore, the beam polarizations can be selected so as to minimize the effect from residual gas atoms (a vacuum of the order of 10^{-10} mm would still be required). While the result still appears to be undetectably small, it may be of sufficient interest to be noted, if only to emphasize how linear the vacuum actually is.

We consider three high-energy pulses of duration τ, containing energy $\mathcal{E}$, each, in a plane at relative angles appropriate to coherent production of a fourth frequency interacting simultaneously in a region of thickness d normal to the propagation plane. The number of ω_4 photons per burst N_4 is then given by

$$N_4=\Gamma(e^2/\hbar c)^4(\mathcal{E}/mc^2)^3(\hbar/mc)^6(1/\lambda_4 d^2c^2\tau^2),$$

where Γ is a geometrical factor of order three. In order to get even a single photon per burst, it is necessary to make very extreme assumptions about the variables. As an example we mention $\mathcal{E}\approx 1$ kJ, $\tau\approx 1/c$, $d\approx\lambda$.