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Electro-optic and piezo-optic tuning of second-order nonlinear processes in crystals

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The power-handling capability of crystals utilized for second harmonic generation (SHG) and other nonlinear frequency conversions has until recently been limited to a few watts of average output power, chiefly because of two conditions, namely, thermal instability and thermal gradients. Now, techniques have been invented that correct these conditions with a resulting order of magnitude increase in the average output power produced by SHG. This paper presents the theory for two techniques that solve the problem of thermal instability, namely, electro-optical tuning (EOT) and piezo-optical tuning (POT). Beam shaping, which prevents thermal gradients, will be treated in a separate paper. In this paper, the general physical theory is discussed both for EOT and POT. The equation for the phase-match condition is given. Interactive effects of temperature change ΔT, the applied electric field (**E**), and the stress field σ on a fixed-position crystal are treated. The general equations for EOT and POT are developed. Examples of the effect on SHG in cesium dideuterium arsenate (CD*A) are given.

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I. INTRODUCTION

The average power outputs of second-order nonlinear processes in crystals, such as parametric amplification and second-harmonic generation (SHG), have for many years been limited to a few watts. Although pump lasers that deliver hundreds of watts are available, the crystals have not been able to handle more than 10–20 W of average power. Efficient conversion by crystals, and hence the delivery of large quantities of average output power, is governed by many factors, such as nonlinear coefficients, peak power density, and beam quality. However, the principal factor that has made it difficult to deliver large average power outputs is the absorption of incident energy by the crystal, which creates two destructive conditions. One is thermal instability, which prevents establishment of the phase-matching condition for longer than brief moments. The other is the presence of thermal gradients, which prevent the phase-matching condition from existing in all parts of the crystal. Typically, depending on the magnitude of the input power, initial conversion efficiencies of 30–50% will in several seconds decrease to ineffective levels.

The problem has been solved by the development of techniques that prevent the development of these destructive conditions. Use of these techniques has resulted in average output of tens of watts.

The technique for eliminating thermal gradients, which involves beam shaping, will be described in a separate paper. This paper deals with the techniques for overcoming the problem of thermal instability. Thermal instability can be eliminated by various means, in particular, by the application of electrical and stress fields. This paper presents a physical theory of electro-optic tuning (EOT) and piezo-optic tuning (POT) for second-order nonlinear processes and gives examples of second-harmonic generation (SHG) in cesium dideuterium arsenate (CD*A).

II. GENERAL THEORY

A. Second-order nonlinear optical process—Phase-match control

While EOT and POT are generally applicable to many optical processes in solids, this discussion is limited to second-order nonlinear optical processes, which are the most important of the nonlinear optical processes.[1,2] Both tuning concepts will be treated in the same discussion here, even though, in practice, they are usually not applied together.

Here, we assume the presence of three quasimonochromatic fields,

$$\mathbf{E}=\mathbf{E}(\omega_1)+\mathbf{E}(\omega_2)+\mathbf{E}(\omega_3), \tag{1}$$

with $\omega_3=\omega_1+\omega_2$. In parametric amplication (or, if feedback is present, parametric oscillation), down-conversion, or difference-frequency generation, ω_3 is the pump. In sum-frequency generation or parametric upconversion (of which SHG is a special case), ω_1 and ω_2 are the pumps. Maxwell's equations of Eq. (1) can be decomposed into three equations nonlinearly coupled with one another through the polarizations,

$$P(\omega_1)=X^{(1)}(\omega_1)\cdot\mathbf{E}(\omega_1)+X^{(2)}:\mathbf{E}(\omega_2)\mathbf{E}(\omega_3),\qquad \text{etc.},$$

where $X^{(1)}$ and $X^{(2)}$ are the first- and second-order polarizabilities.

In the copresence of all three waves, the direction and magnitude of energy flow is dependent[2] on the relative phase relationships among them. Maxwell's equations provide the conditions for optimal energy transfer, in terms of the phase-mismatch quantity $\Delta\mathbf{K}$, defined by

$$\begin{aligned}\Delta\mathbf{K}&=\mathbf{K}(\omega_3)-\mathbf{K}(\omega_2)-\mathbf{K}(\omega_1)\\&=0.\end{aligned} \tag{2}$$

Equation (2) defines the phase-match condition.[1–3]

B. EOT and POT of the phase-matching condition

To produce the phase-match condition necessary for

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a given second-order nonlinear process, it is necessary to make use of the dispersion, birefringence, and temperature conditions of a given crystal. Thus, the most common techniques involve (a) choice of material, (b) angular tuning, and (c) temperature tuning. EOT,[4-7] and now POT, both reported for the first time in this paper, have recently been added to this list of phase-match techniques.

In particular, EOT and POT can be used to compensate for temperature fluctuations in high-average-power applications. For the rest of this section, our discussion will be restricted to the interactions of temperature change ΔT, applied electric field $\mathbf{E}$, and the stress field σ on a given crystal in a fixed position. We assume that at the initial condition of $\Delta T = \mathbf{E} = \sigma = 0$, the temperature of the crystal is in the phase-match condition, which provides the desired second-order nonlinear process in terms of wavelengths, polarizations, and directions or propagation of the participant waves. Thus, any further disruption of the phase-match condition may be described by the phase-mismatch quantity ΔK as

$$\Delta\mathbf{K} = \Delta\mathbf{K}(\omega_3) - \Delta\mathbf{K}(\omega_1) - \Delta\mathbf{K}(\omega_2), \tag{3}$$

where $\Delta\mathbf{K}(\omega_1)$, etc., are the changes in $\mathbf{K}(\omega_1)$, etc., that have been brought about by ΔT, $\mathbf{E}$, and σ.

It is conceivable that judicious application of $\mathbf{E}$ and σ can neutralize or compensate for disruptions caused by crystal heating, ΔT. ΔK [Eq. (3)] would then still be equal to zero; that is,

$$\Delta\mathbf{K} = \Delta\mathbf{K}(\omega_1) - \Delta\mathbf{K}(\omega_2) - \Delta\mathbf{K}(\omega_3) = 0. \tag{4}$$

It is assumed that the parameters ω_1, ω_2, and ω_3, along with their polarizations and directions of propagation, will be the same as in the initial phase-match condition. This compensation by the application of $\mathbf{E}$ and σ is the basis of EOT and POT.

A wave vector is related to the index of refraction by

$$|\mathbf{K}(\omega)| = 2\pi\omega n_{\hat{e}}(\omega)/c, \tag{5}$$

where c is the vacuum speed of light and $n_{\hat{e}}(\omega)$ is the frequency ω and polarization $\hat{e}$ dependent index of refraction, which is generally described by the index ellipsoid

$$B_{ij}(\omega)X_i X_j = 1 \quad (i,j = 1,2,3). \tag{6}$$

The index $n_{\hat{e}}(\omega)$ of an arbitrary polarization $\hat{e} = (e_1, e_2, e_3)$ is given by Eq. (7),

$$n_{\hat{e}}(\omega) = (\textstyle\sum_{ij} B_{ij}\, e_i\, e_j)^{-1/2}. \tag{7}$$

In a principal coordinate system, cross terms of B_{ij} vanish and

$$n_i(\omega) = B_{ii}^{-1/2} \quad (i = 1,2,3). \tag{8}$$

Under the influence of ΔT, $\mathbf{E}$, and σ, the index is still represented by an ellipsoid, although somewhat changed, and Eq. (5) becomes

$$[B_{ij}(\omega) + \Delta B_{ij}(\omega)]X_i X_j = 1, \tag{9}$$

with

$$\Delta B_{ij}(\omega) = \theta_{ijl}(\omega)\Delta T + z_{ijk}(\omega)E_k + \pi_{ijkl}\sigma_{kl} \quad (i, j, k, l = 1, 2, 3), \tag{10}$$

where θ_{ij}, z_{ijk}, and π_{ijkl} are components of the pyro-optic, electro-optic, and piezo-optic constants, which are second-, third, and fourth-rank tensors, respectively.

By using Eqs. (5) and (10) and a differentiation of Eq. (7), Eq. (4) can be rewritten as

$$\begin{aligned}\Delta K = &\{[-\pi\omega_3 n_{\hat{e}}^3(\omega_3)/c] \\ &\times e_i(\omega_3)e_j(\omega_3)[\theta_{ij}(\omega_3)\Delta T + z_{ijk}(\omega_3)E_k + \pi_{ijlm}(\omega_3)\sigma_{lm}]\} \\ &-\{\text{similar expression for } \omega_1\} \\ &-\{\text{similar expression for } \omega_2\} \\ = &\ 0.\end{aligned} \tag{11}$$

This is the general equation for the EOT and POT of second-order nonlinear processes for the compensation of temperature fluctuations. It usually reduces to a simpler form in specific applications.

C. Genuine and para-EOT or para-POT

There are cases when the applied $\mathbf{E}$ and σ may cause such a change to the index ellipsoid that one or more of the originally defined interacting beams simply must undergo change, e.g., polarization, during propagation, in ways directly and adversely affecting the nonlinear process itself. In general, the deleterious effects can be made arbitrarily small by limiting the magnitudes of the applied fields. In fact, in many cases, the deleterious effects are found to be negligibly small when fields capable of producing ample tuning range are used. That is, EOT and/or POT and the present theory are still applicable for all practical purposes. Nevertheless, such cases are called "para"-EOT or "para-POT, as contrasted to "genuine" EOT or POT, because the applied electric or stress field never completely compensates for a shift in temperature in the sense of Eq. (4).

III. EXAMPLE—SECOND-HARMONIC GENERATION IN CD*A

We are now ready to look at a special case of EOT and POT, namely, their application to SHG in temperature-tuned 90°-phase-matched CD*A. As noted, SHG is a special case of parametric up-conversion where

$$\omega_1 = \omega_2 \equiv \omega$$

is the pump, and (12)

$$\omega_3 = 2\omega$$

is the product. The relative SHG efficiency is given by the familiar equation[4]

$$P(2\omega)/[P(2\omega)]_{\max} = \sin^2(\tfrac{1}{2}\Delta KL)/(\tfrac{1}{2}\Delta KL)^2, \tag{13}$$

where L is the length of crystal and

$$\Delta K = \Delta K(2\omega) - 2\Delta K(\omega), \tag{14}$$

which is reduced from Eq. (3) for the present case of type-I phase matching.

An isomorph of KDP, CD*A has a $\bar{4}2m$ point group symmetry. The optimal SHG orientation is shown in Fig. 1. In temperature-tuned 90° phase matching, the normally degenerate x_1 and x_2 axes are at 45° with the propagation direction $\mathbf{k}$ of the 1.06-μ fundamental beam

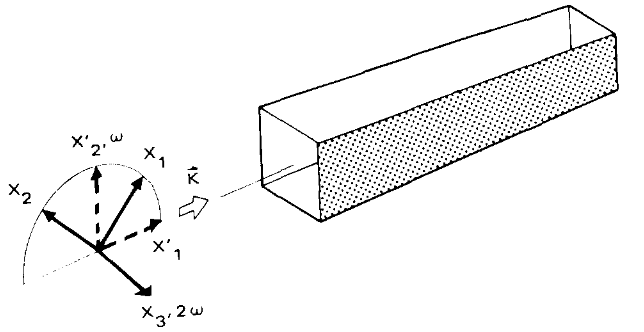

FIG. 1. Crystal orientation of CD*A in SHG. The fundamental $1.06\text{-}\mu\ \omega$ is polarized in the degenerate x_1x_2 (or xy) plane at 45° with either axis. The green light 2ω produced by SHG is polarized along the x_3 (or a or c) axis.

which is polarized in the x_1x_2 plane. The $0.53\text{-}\mu$ SHG beam is polarized along the x_3 axis. The polarization unit vectors are therefore

$$\hat{e}(2\omega) = (\sqrt{2})^{-1}(\hat{x}_1 + \hat{x}_2)$$

and (15)

$$\hat{e}(\omega) = \hat{x}_3.$$

Substitution of Eqs. (12) and (15) into the general equation (11), and noting that $n_e(2\omega) = n_e(\omega) = n$ by definition of phase matching, yields

$$\{\tfrac{1}{2}[\theta_{11}(2\omega) + \theta_{22}(2\omega) + \theta_{12}(2\omega) + \theta_{21}(2\omega)] - \theta_{33}(\omega)\}\Delta T + \{\tfrac{1}{2}[z_{11k}(2\omega) + z_{22k}(2\omega) + z_{12k}(2\omega) + z_{21k}(2\omega)] - z_{33k}(\omega)\}E_k + \{\tfrac{1}{2}[\pi_{11lm}(2\omega) + \pi_{22lm}(2\omega) + \pi_{12lm}(2\omega) + \pi_{21lm}(2\omega)] - \pi_{33lm}(\omega)\}\sigma_{lm}. \quad (16)$$

Further simplification comes from crystal symmetry, which dictates what forms the tensors θ, z, and π must take. For CD*A, in reduced notations, Eq. (10) reads reads[8-10]:

$$\begin{bmatrix}\Delta B_1\\ \Delta B_2\\ \Delta B_3\\ \Delta B_4\\ \Delta B_5\\ \Delta B_6\end{bmatrix} = \begin{bmatrix}\bullet\\ \bullet\\ \bullet\\ \cdot\\ \cdot\\ \cdot\end{bmatrix}\Delta T + \begin{bmatrix}\cdot & \cdot & \cdot\\ \cdot & \cdot & \cdot\\ \cdot & \cdot & \cdot\\ \bullet & \cdot & \cdot\\ \cdot & \bullet & \cdot\\ \cdot & \cdot & \bullet\end{bmatrix}\begin{bmatrix}E_1\\ E_2\\ E_3\end{bmatrix} + \begin{bmatrix}\bullet & \bullet & \bullet & \cdot & \cdot & \cdot\\ \bullet & \bullet & \bullet & \cdot & \cdot & \cdot\\ \bullet & \bullet & \bullet & \cdot & \cdot & \cdot\\ \cdot & \cdot & \cdot & \bullet & \cdot & \cdot\\ \cdot & \cdot & \cdot & \cdot & \bullet & \cdot\\ \cdot & \cdot & \cdot & \cdot & \cdot & \bullet\end{bmatrix}\begin{bmatrix}\sigma_1\\ \sigma_2\\ \sigma_3\\ \sigma_4\\ \sigma_5\\ \sigma_6\end{bmatrix}. \quad (17)$$

(Small dots = 0, connected large dots have equal values.)

Thus, Eq. (16) simplifies to

$$[\theta_1(2\omega) - \theta_3(\omega)]\Delta T + z_{6k}(2\omega)E_k + [\pi'_{1l}(2\omega) - \pi'_{6l}(2\omega) - \pi'_{3l}(\omega)]\sigma_l,$$
$$k = 1,\ 2,\ 3; \quad l = 1, \ldots, 6, \quad (18)$$

where

$$\pi'_{il} = \pi_{il} \quad \text{if } l = 1,\ 2,\ 3$$
$$= \tfrac{1}{2}\pi_{il} \quad \text{if } l = 4,\ 5,\ 6,$$

due to the definition of the reduced notation for the π tensor.

A. Electro-optic tuning

For EOT, $\sigma_l = 0$. According to Fig. 1, the applied field $\mathbf{E}$ may conveniently take the form of either $\mathbf{E} = E\hat{x}_3$ or $\mathbf{E} = (E/\sqrt{2})(\hat{x}_1 + \hat{x}_2)$. (Other crystal orientations are possible but they will not be discussed here.)

1. EOT case I

In case I, $E = Ex_3$. Eq. (18) becomes

$$[\theta_1(2\omega) - \theta_3(\omega)]\Delta T + z_{63}(2\omega)E = 0;$$

and, it follows that

$$(\Delta T/E)_{\text{I}} = z_{63}(2\omega)/[\theta_1(2\omega) - \theta_3(w)],$$

which is the case-I EOT coefficient, which gives the shift in the phase-match temperature due to an applied E field (see Fig. 2). Equivalently,

$$(\Delta T/E)_{\text{I}} = n^3 z_{63}(2\omega)\left(2\frac{d}{dT}[n_1(2\omega) - n_3(\omega)]\right)^{-1}, \quad (19)$$

the last step being a simple application of Eqs. (8) and (10).

However, a more useful expression which is in terms of experimental observables is

$$(\Delta T/E)_I = Ln^3\delta T z_{63}(2\omega)/\lambda(\omega), \quad (20)$$

where $\lambda(\omega)$ is the vacuum wavelength of the fundamental beam and δT is the distance between the maximum and the first minimum on the SHG output versus temperature curve which is shown in Fig. 3. (It is also a full width at 0.405 maximum). Equation (20) was discussed in paper on EOT[4] and can easily be obtained by (i) noting that Fig. 2 is described by Eq. (12) which has its first minimum when $\frac{1}{2}\Delta kL = \pi$, and by (ii) applying Eq. (5) to Eq. (19).

To check if this case is a genuine or a para-EOT, the electro-optic effect on propagative beams ω and 2ω should be scrutinized. According to Eq. (17)

$$\begin{bmatrix}\Delta B_1\\ \Delta B_2\\ \Delta B_3\\ \Delta B_4\\ \Delta B_5\\ \Delta B_6\end{bmatrix} = E\begin{bmatrix}\cdot & \cdot & \cdot\\ \cdot & \cdot & \cdot\\ \cdot & \cdot & \cdot\\ \bullet & \cdot & \cdot\\ \cdot & \bullet & \cdot\\ \cdot & \cdot & \bullet\end{bmatrix}\begin{bmatrix}\cdot\\ \cdot\\ 1\end{bmatrix} = E\begin{bmatrix}\cdot\\ \cdot\\ \cdot\\ \cdot\\ \cdot\\ z_{63}\end{bmatrix}$$

The cross term $\Delta B_6 = z_{63}$ means for the index ellipsoid, a 45° rotation of the originally degenerate $x_1 - x_2$ axes to the nondegenerate $x'_1 - x'_2$ axes as shown in Fig. 1.

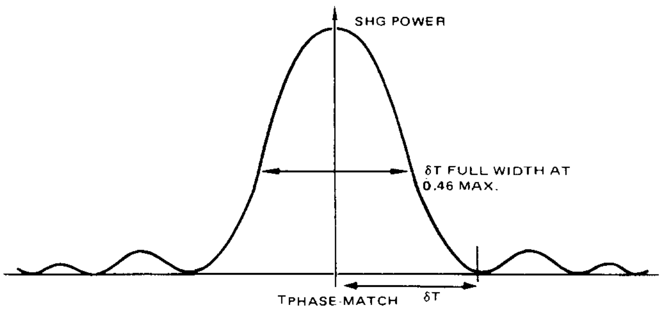

FIG. 2. Phase-match temperature profile.

This leaves both the ω and 2ω beams still polarized along principal axes throughout the crystal. Only thin velocities are electro-optically modified. It is theoretically possible to find an E field of appropriate magnitude to compensate for any temperature change in the sense of Eq. (4); no degradation in the overall FD efficiency will result. This is a case of genuine EOT.

2. EOT case II

In case II, where $\mathbf{E}=(E/\sqrt{2})(\hat{x}_1+\hat{x}_2)$, Eq. (18) be becomes

$$[\theta_1(2\omega)-\theta_3(\omega)]\Delta T+(E/\sqrt{2})[z_{61}(2\omega)+z_{62}(2\omega)]=0. \qquad (21)$$

Since both z_{61} and z_{62} vanish according to Eq. (17), there is no EOT. In any event, this case would not have been a genuine EOT, for the electro-optic effect here is, according to Eq. (17),

$$\begin{bmatrix}\Delta B_1\\ \Delta B_2\\ \Delta B_3\\ \Delta B_4\\ \Delta B_5\\ \Delta B_6\end{bmatrix}=\frac{E}{\sqrt{2}}\begin{bmatrix}\cdot&\cdot&\cdot\\ \cdot&\cdot&\circ\\ \cdot&\circ&\cdot\\ \bullet&\cdot&\cdot\\ \cdot&\bullet&\cdot\\ \cdot&\cdot&\bullet\end{bmatrix}\begin{bmatrix}1\\1\\ \cdot\end{bmatrix}=\frac{E}{\sqrt{2}}\begin{bmatrix}\cdot\\ \cdot\\ \cdot\\ z_{41}\\ z_{41}\\ \circ\end{bmatrix} \qquad (22)$$

The creation of the cross terms ΔB_4 and ΔB_5 implies a rotation of all three principal axes of the index ellipsoid. Both ω and 2ω beams, as defined in the original phase-match condition, must propagate with the polarization vectors gradually changing, resulting in a disruption of the SHG process itself. So, even if EOT had existed according to Eq. (21), this case would have been one of para-EOT at best.

B. Piezo-optic tuning

For POT, in Eq. (18) $E_k=0$. Two directions of stress can be conveniently applied, namely, along $\hat{x}_3$ and $(\hat{x}_1+\hat{x}_2)$.

1. POT case I

For case I, $\sigma=(1,0,0,0,0,0)/\sqrt{3}$. Upon using the symmetry rules for tensor in Eq. (17), Eq. (18) becomes

$$[\theta_1(2\omega)-\theta_3(\omega)]\Delta T+[\pi_{13}(2\omega)-\pi_{33}(\omega)]\sigma=0. \qquad (23)$$

For case I, the POT coefficient $(\Delta T/\sigma)_{\mathrm{I}}$, which describes the shift in the phase-match temperature due to an applied stress σ, is

$$(\Delta T/\sigma)_{\mathrm{I}}=[\pi_{33}(\omega)-\pi_{13}(2\omega)][\theta_1(2\omega)-\theta_3(\omega)]^{-1} \qquad (24)$$

It can easily be shown that

$$(\Delta T/\sigma)_{\mathrm{I}}=[\pi_{33}(\omega)-\pi_{13}(2\omega)]Ln^3\delta T/\lambda(\omega) \qquad (25)$$

by reasons identical to those leading to Eq. (20).

Is this a case of genuine POT? According to Eq. (17), the piezo-optic effect is

$$\begin{bmatrix}\Delta B_1\\ \Delta B_2\\ \Delta B_3\\ \Delta B_4\\ \Delta B_5\\ \Delta B_6\end{bmatrix}=\sigma\begin{bmatrix}\bullet&\bullet&\bullet&\cdot&\cdot&\cdot\\ \bullet&\bullet&\bullet&\circ&\cdot&\cdot\\ \bullet&\bullet&\bullet&\cdot&\cdot&\circ\\ \cdot&\cdot&\cdot&\bullet&\cdot&\cdot\\ \cdot&\cdot&\cdot&\cdot&\bullet&\circ\\ \cdot&\cdot&\cdot&\cdot&\circ&\bullet\end{bmatrix}\begin{bmatrix}\cdot\\ \cdot\\ 1\\ \circ\\ \cdot\\ \cdot\end{bmatrix}=\sigma\begin{bmatrix}\pi_{13}\\ \pi_{13}\\ \pi_{33}\\ \cdot\\ \cdot\\ \cdot\end{bmatrix}$$

Evidently the crystal symmetry, and thus the principal axes of the index ellipsoid, is unchanged. It is a case of genuine POT.

2. POT case II

In case II, where the applied stress is parallel to $(\hat{x}_1+\hat{x}_2)$,

$$\sigma=(\sigma/\sqrt{2})(1,1,0,0,0,2)$$

in the reduced notation.[11] Upon application of the symmetry rules for CD*A σ tensor in Eq. (17), Eq. (18) becomes

$$[\theta_1(2\omega)-\theta_3(\omega)]\Delta T+[\pi_{11}(2\omega)-2\pi_{31}(\omega)+\pi_{12}(2\omega)+\tfrac{1}{2}\pi_{66}(2\omega)](\sigma/\sqrt{2})=0.$$

It follows that,

$$\begin{aligned}(\Delta T/\sigma)_{\mathrm{II}}&=[2\pi_{31}(\omega)-\pi_{11}(2\omega)-\pi_{12}(2\omega)-\tfrac{1}{2}\pi_{66}(2\omega)]\\&\quad\times[\theta_1(2\omega)-\theta_3(\omega)]^{-1}\\&=[2\pi_{31}(\omega)-\pi_{11}(2\omega)-\pi_{12}(2\omega)-\tfrac{1}{2}\pi_{66}(2\omega)]\\&\quad\times Ln^3\delta T/\lambda(\omega)\end{aligned} \qquad (26)$$

for the POT coefficient in case II. Again, is this a genuine case of POT? According to Eq. (17), the piezo-optic effect is given by

$$\begin{bmatrix}\Delta B_1\\ \Delta B_2\\ \Delta B_3\\ \Delta B_4\\ \Delta B_5\\ \Delta B_6\end{bmatrix}=\frac{\sigma}{\sqrt{2}}\begin{bmatrix}\bullet&\bullet&\bullet&\cdot&\cdot&\cdot\\ \bullet&\bullet&\bullet&\cdot&\cdot&\circ\\ \bullet&\bullet&\bullet&\cdot&\cdot&\cdot\\ \cdot&\cdot&\cdot&\bullet&\cdot&\cdot\\ \cdot&\cdot&\cdot&\cdot&\bullet&\cdot\\ \cdot&\cdot&\cdot&\cdot&\cdot&\bullet\end{bmatrix}\begin{bmatrix}1\\ 1\\ \cdot\\ \cdot\\ \circ\\ 2\end{bmatrix}$$

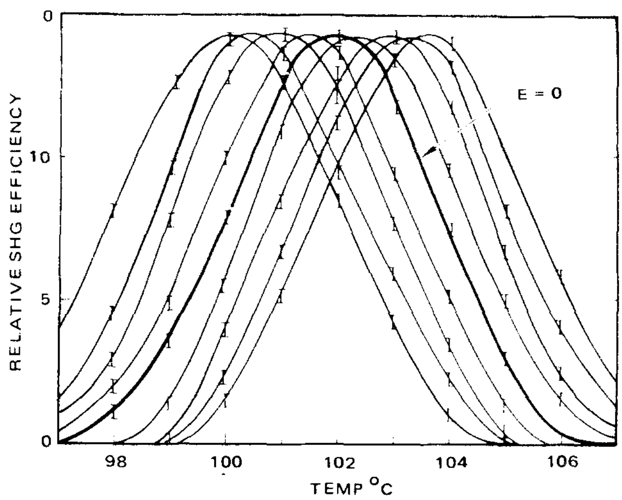

FIG. 3. Unclamped electro-optic effects on SHG. Efficiency versus temperature for different applied E fields. E-field pulse duration $\sim 200\ \mu s$, $\Delta E = \pm 850$, ± 1800, ± 2750, and ± 3250 V/cm.

$$= \frac{\sigma}{\sqrt{2}} \begin{bmatrix} \pi_{11} + \pi_{22} \\ \pi_{11} + \pi_{12} \\ 2\pi_{13} \\ \cdot \\ \cdot \\ 2\pi_{66} \end{bmatrix} .$$

This is similar to EOT case II. The originally degenerate $x_1 - x_2$ axis of the index ellipsoid is rotated 45° to the nondegenerate $x_1' - x_2'$ axis. n_3 also changes due to $\Delta B_3 = 2\pi_{13}$; but the ω and 2ω waves are still polarized along principal axes, x_2' and x_3, respectively, throughout the crystal. So, this is also a genuine POT.

3. POT by hydrostatic pressure

Conceivably, hydrostatic pressure can be used for POT when the crystal is submerged in a fluid. For some applications, this technique may offer practical advantages over unidirectional POT. Here, Eq. (17) reads

$$\begin{bmatrix} \Delta B_1 \\ \Delta B_2 \\ \Delta B_3 \\ \Delta B_4 \\ \Delta B_5 \\ \Delta B_6 \end{bmatrix} = \sigma \begin{bmatrix} \bullet & \bullet & \bullet & \cdot & \cdot & \cdot \\ \bullet & \bullet & \bullet & \cdot & \cdot & \cdot \\ \bullet & \bullet & \bullet & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \bullet & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \bullet & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \bullet \end{bmatrix} \begin{bmatrix} 1 \\ 1 \\ 1 \\ \cdot \\ \cdot \\ \cdot \end{bmatrix}$$

$$= \sigma \begin{bmatrix} \pi_{11} + \pi_{12} + \pi_{13} \\ \pi_{11} + \pi_{12} + \pi_{13} \\ 2\pi_{31} + \pi_{33} \\ \cdot \\ \cdot \\ \cdot \end{bmatrix} .$$

The equivalent of Eqs. (24) and (25) for hydrostatic POT is

$$(\Delta T/\sigma)_H = [2\pi_{31}(\omega) + \pi_{33}(\omega) - \pi_{11}(2\omega) - \pi_{12}(2\omega) - \pi_{13}(2\omega)]\, L n^3 \delta T/\lambda(\omega).$$

As hydrostatic pressure never changes the symmetry of a crystal, only genuine POT can occur.

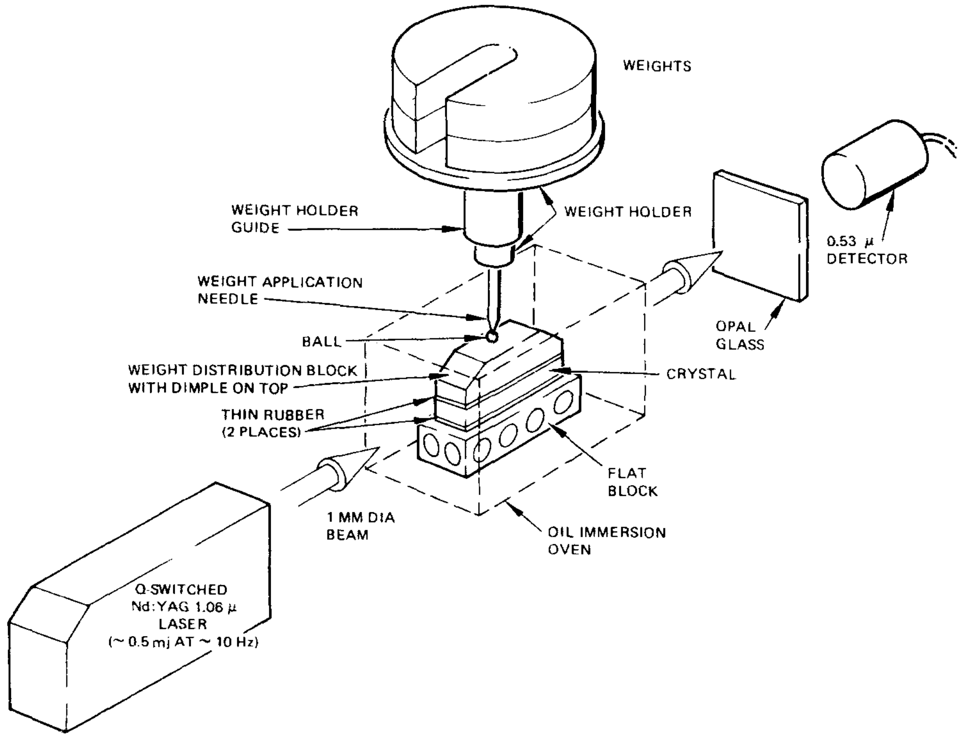

FIG. 4. Experimental setup for piezo-optic tuning (POT).

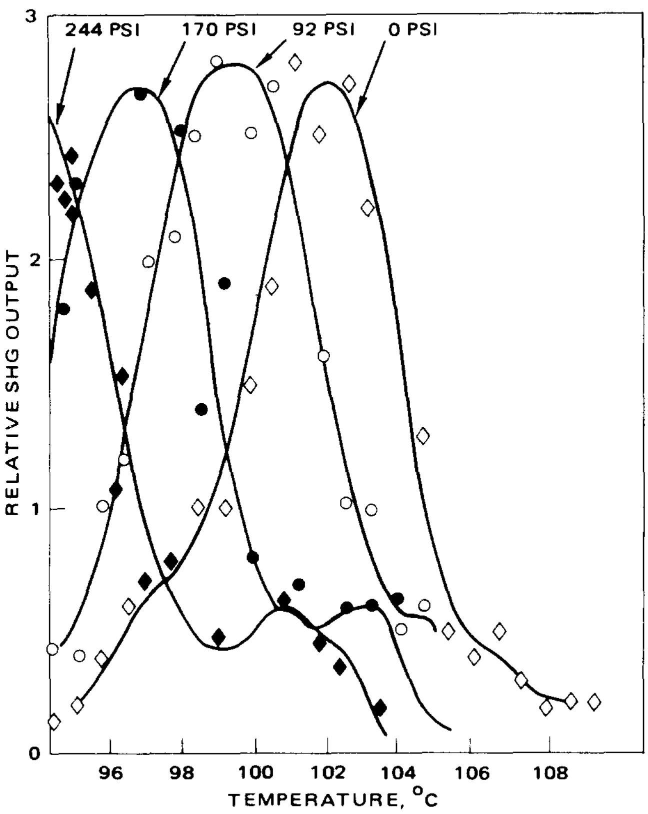

FIG. 5. Piezo-optic tuning (POT) in Cd*A. Relative SHG output versus temperature for various pressures for pressure along $(\hat{x}_1+\hat{x}_2)$.

V. EXPERIMENTAL RESULTS

Experiments involving the EOT for SHG in CD*A have been reported in Ref. 4. Therefore, only the principal result will be reproduced here. Figure 3 shows the SHG power versus temperature for various applied E fields.

With respect to POT, Fig. 4 shows the experimental setup that was used for applying unilateral pressure to the crystal while maintaining it at or near the 90°-phase-match temperature. The oil immersion oven was rigidly mounted to the optical bench to ensure that the crystal remained immobile while the pressure was applied. The 1-mm-diam 1.06-μm beam from a 20-mJ/10-Hz YAG laser traversed the center of the $0.4\times0.4\times1\text{-cm}^3$ crystal. Monitoring of the position of surface reflections verified that the crystal did not move.

Several precautions were taken to ensure that pressure was applied uniformly. A massive weight holder and guide directed the force of the weights onto the crystal. The surfaces of the crystal were ground flat and parallel. When mounted, the crystal, cushioned by thin slips of paper, was sandwiched between two flat steel blocks. The upper block had a dimple in its center. This dimple received the steel ball embedded in the tip of the weight application needle.

The tip of the needle is partially immersed in the oil, and it is wound with heating coils that maintain its temperature identical to that of the rest of the oven.

TABLE I. Electro-optic and piezo-optic tuning coefficients for

	EOT coefficient $(\Delta T/E)$ (°C cm/V)	POT coefficient $(\Delta T/\sigma)$ [°C cm²/dyn (°C/psi)]
Case I		
E or $\sigma \parallel \hat{x}_3$	$5.5\times10^{-4}\pm6\%$	$0\pm2.8\times10^{-8}$ $(0\pm2\ \ \times10^{-3})$
Case II		
E or $\sigma \parallel (\hat{x}_1+\hat{x}_2)$	$0\pm2\times10^{-5}$	$4.4\ \times10^{-7}\pm10\%$ $(3.12\times10^{-2}\pm10\%)$

These precautions, and the procedure, ensure that the temperature of the crystal is uniformly constant.

The experimental procedure follows: (1) Allow oven to stabilize at a temperature with no weight applied. (2) Take SHG reading. (3) Lower weight(s) and quickly take a second reading. (4) Quickly raise weight, note that reading returns to previous value. (5) Change temperature and repeat steps (1)–(5). Temperature-profile curves for several different pressures, 0, 92, 170, and 244 psi, were plotted from this data, and the shift read off. The results are plotted in Fig. 5. Shifts of as much as 10 °C in the phase-matched temperature were obtained. Figure 5 also shows that large applied pressures can cause some degradation in efficiency and broadening of the SHG curves. This is believed to be due to stress fringe effects in this finite-sized crystal, and not an indication of "para-POT".

The EOT[4] and POT coefficients for SHG in CD*A have been measured, and they are listed in Table I.

The analysis in Sec. III A 2 correctly predicted a zero coefficient for EOT, case II. It also predicted "genuine" tuning and nonzero coefficients for the rest. Surprisingly, the coefficient for POT case I was found to be zero within experimental accuracies. Thus, it follows from Eq. (24) that

$$\pi_{33}(\omega)-\pi_{13}(2\omega)\sim0.$$

For POT case II, the application of Eq. (25) to Fig. 5 yields

$$2\pi_{31}(\omega)-\pi_{11}(2\omega)-\pi_{12}(2\omega)-\tfrac{1}{2}\pi_{66}(2\omega)$$
$$=(2.5\pm0.3)\times10^{-12}\ \text{cm}^2/\text{dyn}, \qquad (26)$$

where the values $L=1$ cm, $n=1.5T=5.5$ °C have been used. There are insufficient published data on the piezo-optic tensor π on this crystal for a corroborating comparison, even though the order of magnitude of Eq. (26) appears correct.

However, in EOT case I, the application of Eq. (20) in Ref. 4 resulted in the value of the electro-optic constant $z_{63}(2\omega)=(24.3\pm0.6)\times10^{-10}$ cm/V, which compares well with extrapolations of published values. It can be seen from Fig. 4 that no detectable degradation of SHG efficiency occurred as a result of the applied E field which is consistent with the fact that this is genuine EOT. The E-field fringe effect is believed to be small due to the relatively low resistivity of CD*A crystal, about $4\times10^5\ \Omega$ cm.

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[9]Algebraically, Eq. (17) reads $\Delta B_i = \theta_i \Delta T + z_{ij} E_j + \pi_{ik} \sigma_k$ with i, $k = 1, \ldots, 6$, and $j = 1, 2, 3$. Note that (i) $\Delta B_4 = \Delta B_{32} = \Delta B_{23}$, etc., (ii) in changing from z_{ijk} to z_{ij} no factors of $\frac{1}{2}$ or 2 appear (iii) $\pi_{mn} = \pi_{ijkl}$ when $n = 1, 2$, or 3: $\pi_{mn} = 2\pi_{ijkl}$ when $n = 4, 5$, or 6, and (iv) $\sigma_4 = \sigma_{32} = \sigma_{23}$, etc.
[10]θ takes this from because temperature change does not alter the symmetry of the crystal and because CD*A has a uniaxial symmetry.
[11]This can be obtained by observing that this stress tensor is $(1, 0, 0, 0, 0, 0)$ in the coordinate system $[\hat{x}_1', \hat{x}_2', \hat{x}_3'] = [(1/\sqrt{2})(\hat{x}_1 + \hat{x}_2), (1/\sqrt{2})(\hat{x}_1 - \hat{x}_2), \hat{x}_3]$. When this coordinate is rotated to the coordinate $(\hat{x}_1, \hat{x}_2, \hat{x}_3)$, the stress tensor takes the form as indicated.