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Influence of piezoelectricity on the photorefractive effect

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We demonstrate that the spatially modulated electric field that is associated with a photorefractive grating generates stress and strain components with symmetries that are different from those induced by a uniform electric field. Therefore, because of piezoelectricity and the elasto-optic effect, the symmetries of the effective dielectric and electro-optic constants to be used to describe the photorefractive effect differ from the symmetries of the usual dielectric and electro-optic tensors. We derive analytical expressions to be used to compute these new constants from the measured clamped and unclamped dielectric and electro-optic coefficients. Experimental evidence is presented.

1. INTRODUCTION

The physics of the photorefractive effect is now well understood.¹ A photorefractive grating results from a photoinduced charge redistribution in the bulk of the material. The associated photoinduced electric field in turn generates an index change. To predict the kinetics and the steady state of the photorefractive effect, it is therefore necessary to determine the exact relations, first, between the charge pattern and the electric field and, second, between the electric field and the index grating. Because of the elasto-optic effect, the deformations induced by the electric field (piezoelectricity²) must be taken into account. For example, Stepanoy *et al.*³ demonstrated the influence of piezoelectricity (by means of the elasto-optic effect) on the optical index changes in cubic crystals. The dielectric tensor and electro-optic tensor have already been measured for most photorefractive materials and for both clamped and unclamped samples. However, these results, obtained by applying a uniform electric field to the crystal, cannot be directly used for spatially modulated electric fields such as those associated with photorefractive gratings. Here we explain that the deformations induced by spatially modulated electric fields indeed necessarily differ from those induced by uniform fields. We demonstrate that the effective dielectric and electro-optic coefficients that should be used in photorefractive experiments are neither the clamped nor the unclamped coefficients determined by the usual measurements but that they can be computed from these values.

The notation that we need for the mathematical analysis is given in Section 2. In Section 3 we recall the well-known results concerning the refractive index and dielectric changes induced by a uniform electric field. In Section 4 we explain the mechanism of the breaking of the crystal symmetry by a spatially modulated electric field. Then we derive the expressions for the effective dielectric and electro-optic coefficients, and we apply these results to BaTiO₃ and Bi₁₂GeO₂₀ crystals.

2. BASIC NOTATION

An internal electric field \mathbf{E} inside a piezoelectric crystal modifies the stress σ , the strain δ , and the electric dis-

placement \mathbf{D} . We may choose at least two different equivalent sets of independent variables.² In what follows we use (σ, \mathbf{E}) and (δ, \mathbf{E}) . With the Einstein summation convention we have

$$\delta_{ij} = s_{ijkl}\sigma_{kl} + d_{kij}E_k, \quad (1a)$$

$$D_i = d_{ijk}\sigma_{jk} + \epsilon_0\epsilon_{ij}^{\sigma=0}E_j, \quad (1b)$$

or

$$\sigma_{ij} = c_{ijkl}\delta_{kl} - e_{kij}E_k, \quad (2a)$$

$$D_i = e_{ijk}\delta_{jk} + \epsilon_0\epsilon_{ij}^{\delta=0}E_j, \quad (2b)$$

where

δ_{ij} , σ_{ij} , E_i , and D_i are the components of the strain tensor, stress tensor, electric field, and electric displacement;

s_{ijkl} and c_{ijkl} are the components of the elastic tensor and rigidity tensor;

e_{ijk} and d_{ijk} are the components of the piezoelectric tensors;

ϵ_0 is the free-space permittivity, and $\epsilon_{ij}^{\delta=0}$ and $\epsilon_{ij}^{\sigma=0}$ are the components of the relative static dielectric tensors for totally clamped (strain-free) and totally unclamped (stress-free) crystals.

Equations (1) and (2) are equivalent, and the relations between their coefficients can be easily determined.² Quite often Eqs. (1) and (2) are expressed with abbreviated notation for simplicity. However, this abbreviated notation is not convenient when changes of axes are needed, and therefore we use it not to carry out the calculation but only to give the numerical values of coefficients.

The effect of the internal electric field \mathbf{E} and of the induced strain δ on the impermeability is given by the change in the impermeability tensor, which has components $\Delta\eta_{ij}$:

$$\Delta\eta_{ij} = r_{ijk}^{\delta=0}E_k + p_{ijkl}^{\sigma=0}\delta_{kl}, \quad (3)$$

with $r_{ijk}^{\delta=0}$ the components of the totally clamped electro-optic tensor and $p_{ijkl}^{\sigma=0}$ the components of the elasto-optic tensor.

The nonvanishing values for the various coefficients of a number of crystals are listed in Appendix A by using abbreviated notations.² The electro-optic coefficients given in Appendix A are the totally unclamped coefficients.

In what follows, given the electric field, we calculate the components $\Delta\eta_{ij}$ and D_i . We first determine the strain δ .

3. UNIFORM INTERNAL ELECTRIC FIELDS

In this section we assume that a given voltage is applied to the sample, resulting in a uniform internal field. d is the spacing between the two parallel electrodes, the modulus of the internal field is V/d , and its direction is perpendicular to the electrodes. We also assume that the strain and the stress are uniform, so that their values inside the material are equal to their values on the boundaries. If the sample is totally unclamped (e.g., $\sigma_{ij} = 0$), the strain components induced by the electric field are given by Eq. (1a). Inserting their values into Eq. (2b) and comparing the result to Eq. (1b), we get

$$\epsilon_{ij}^{\sigma=0} = \epsilon_{ij}^{\delta=0} + e_{ikl}d_{jkl}/\epsilon_0. \quad (4)$$

Similarly, we derive the expression for the totally unclamped electro-optic coefficients $r_{ijk}^{\sigma=0}$ by using Eq. (3):

$$r_{ijk}^{\sigma=0} = r_{ijk}^{\delta=0} + p_{ijlm}d_{klm}. \quad (5)$$

We note that the totally clamped coefficients given in Eqs. (4) and (5) do not correspond to the so-called clamped coefficients that are measured in some experiments where only the deformation along the field axis is forbidden; in the above expressions all six possible deformations are forbidden. We computed the differences between strain-free and stress-free coefficients according to the values listed in Appendix A.

We found that for BaTiO₃

$$\epsilon_1^{\sigma=0} - \epsilon_1^{\delta=0} = 2600, \quad \epsilon_3^{\sigma=0} - \epsilon_3^{\delta=0} = 45,$$

for Bi₁₂GeO₂₀

$$\epsilon_1^{\sigma=0} - \epsilon_1^{\delta=0} = 4.3, \quad r_{41}^{\sigma=0} - r_{41}^{\delta=0} = 0.34 \text{ pm/V},$$

and for GaAs

$$\epsilon_1^{\sigma=0} - \epsilon_1^{\delta=0} = 5 \times 10^{-2}, \quad r_{41}^{\sigma=0} - r_{41}^{\delta=0} = 0.19 \text{ pm/V}.$$

4. SPATIALLY MODULATED INTERNAL ELECTRIC FIELDS

A. Determination of the Strain Components

The above analysis is no longer possible when the electric field is nonuniform. Equations (1) and (2) are not sufficient to determine the deformation at every location inside the material. They give only a relation between the components of δ , σ , \mathbf{E} , and \mathbf{D} . For example, in a cubic material (23 or $\bar{4}3m$), a simple (and wrong) analysis of the symmetries (see Appendix A) seems to indicate that if a field grating is along the x crystallographic axis then only δ_{yz} , δ_{zy} , σ_{yz} , and σ_{zy} can be different from zero. Actually shear deformations (induced by δ_{yz} and δ_{zy}) are spatially modulated along the same axis as the internal field. The macroscopic deformation² along the z axis being

$$U_z = \int \delta_{xx}dx + \delta_{yy}dy + \delta_{zz}dz,$$

the strain components δ_{xx} and δ_{zz} , equal to $\partial U_z/\partial x$, are therefore also not equal to zero (as is true for many others). The relations among δ_{xz} , δ_{zx} , σ_{xz} , and σ_{xy} are still given by Eqs. (1a) and (1b). This nonlocal effect is general for all point groups and cannot be predicted from Eqs. (1) and (2) alone.

To complete Eqs. (1) or (2), we can use the first principle of dynamics.⁴ The forces that are due to stress vanish if the sample is at equilibrium. For a unit volume the expressions of their components are²

$$F_i = \frac{\partial \sigma_{ij}}{\partial x_j} = 0. \quad (6)$$

The boundary conditions impose the values of σ_{ij} on the crystal faces. Equation (6) is valid as soon as the photorefractive buildup time constant τ_g is much larger than the characteristic time constant of the mechanical vibration. If Λ is the grating fringe spacing and if v is the velocity of sound, then this condition can be expressed by

$$\tau_g \gg \Lambda/v. \quad (7)$$

The velocity of sound being equal to a few kilometers per second and the fringe spacing being equal to a few micrometers, τ_g must be greater than a few nanoseconds. Therefore condition (7) is fulfilled in most experiments.

We must remark that, even in the absence of external forces, Eq. (6) does not imply that the stress components σ_{ij} are equal to zero inside the sample. In the more general case strain and stress depend on the three space coordinates. We now restrict our analysis to quasi-infinite crystals whose dimensions are much larger than the fringe spacing Λ of the induced grating. This situation corresponds to most photorefractive experiments, in which the fringe spacing is a few micrometers and the sample dimensions are a few millimeters. However, our analysis does not apply to thin samples, where surface deformations are of importance.⁵ For infinite samples the symmetry implies that the deformation is possible only along the grating wave vector,⁶ which is also the field axis. We thus conduct the calculation for a set of axes in which the first axis x' is parallel to the electric field. All the components in this new set are denoted by a prime. The space derivatives with respect to y' and z' vanish, and Eq. (6) reduces to

$$\frac{\partial \sigma'_{x'x'}}{\partial x'} = 0. \quad (8)$$

As a consequence, the modulated part of $\sigma'_{x'x'}$ equals zero. From Eq. (2) we immediately obtain the only nonzero strain component $\delta'_{x'x'}$:

$$\delta'_{x'x'} = \frac{e'_{x'x'x'}}{c'_{x'x'x'}} E, \quad (9)$$

$$\delta'_{x'y'} = \delta'_{x'z'} = \delta'_{y'z'} = \delta'_{y'y'} = \delta'_{z'z'} = 0,$$

where E denotes the modulus of the field \mathbf{E} .

If a_i denotes the direction cosines of the electric field \mathbf{E} in the crystallographic axes, then $\delta'_{x'x'}$ is expressed as a

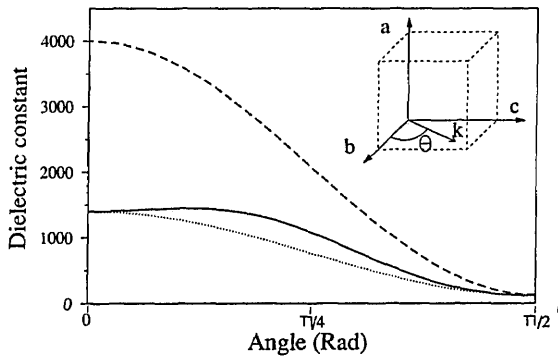


Fig. 1. Effective dielectric constant for BaTiO₃ versus the angle θ between the b crystallographic axis and the direction k of the internal electric field, k being in the (b,c) plane: solid curve, spatially modulated field; dashed curve, uniform field and stress-free sample; dotted curve, uniform field and strain-free sample.

function of the piezoelectric and rigidity tensor components given in those axes:

$$\delta'_{xx'} = HE, \tag{10}$$

with

$$H = \frac{\sum_{i,j,k=1}^3 a_i a_j a_k e_{ijk}}{\sum_{l,m,n,o=1}^3 a_l a_m a_n a_o c_{lmno}}. \tag{11}$$

The strain components can also be expressed relative to the crystallographic axes:

$$\delta_{ij} = a_i a_j HE. \tag{12}$$

Equation (12) demonstrates that a photorefractive grating can generate strain components that cannot be induced by a uniform electric field (and conversely a uniform field can induce strain components that cannot be created by a photorefractive grating). For instance, for cubic crystals and according to Eq. (1a), with the d_{ijk} coefficients given in Appendix A, a spatially uniform electric field along the [111] crystallographic direction can induce only shear deformations (δ_{ij} with $i \neq j$). However, if the field is spatially modulated then all the strain components are equal and differ from zero [Eq. (12)].

We can thus conclude that the piezoelectric tensor symmetry is modified by the inhomogeneous field.

B. Determination of the Effective Dielectric Constant

We derive the relative dielectric tensor ϵ^{pr} , to be used when the electric field is spatially modulated, by inserting Eq. (12) for δ_{ij} into Eq. (2b). We get

$$\epsilon_{ij}^{pr} = \epsilon_{ij}^{\delta=0} + e_{ikj} a_k \frac{H}{\epsilon_0}. \tag{13}$$

Equation (13) is not convenient for performing the calculation and can be misleading because ϵ^{pr} itself depends on the electric field by means of its direction cosines. Poisson's law, which links the modulated electric charge grating to the electric-field displacement, is used to define an effective photorefractive static dielectric constant ϵ_{eff}^{pr} :

$$\text{div}(\epsilon^{pr} \mathbf{E}) = \epsilon_{eff}^{pr} \text{div} \mathbf{E}. \tag{14}$$

From Eqs. (13) and (14) we immediately get

$$\epsilon_{eff}^{pr} = a_i \left(\epsilon_{ij}^{\delta=0} + e_{ikj} a_k \frac{H}{\epsilon_0} \right) a_j. \tag{15}$$

C. Effective Electro-optic Coefficient

In the crystallographic axes the coupling of two optical waves incident upon a photorefractive grating is related to the nonlinear dielectric tensor for optical frequencies:

$$\Delta \epsilon_{ij}^{\omega} = -n_i^2 n_j^2 (\Delta \eta_{ij} / \epsilon_0), \tag{16}$$

where the values $n_i^2 = 1/\epsilon_{ii}^{\omega}$ are the squares of the optical refractive indices.

v_i and w_i being the components of the two optical wave polarization vectors, the coupling strength is proportional

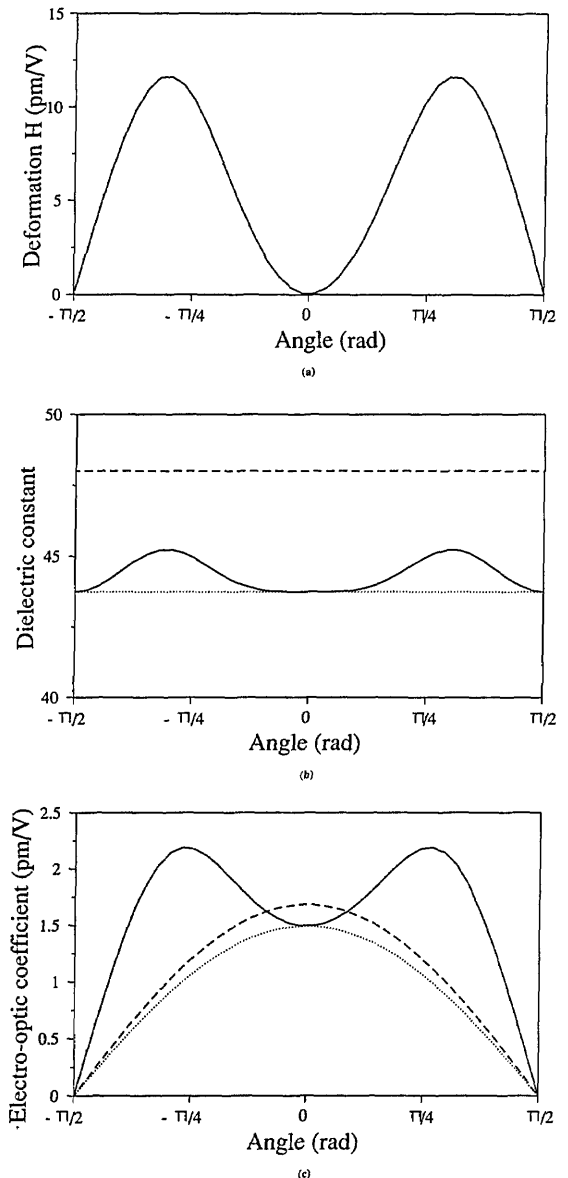


Fig. 2. Dependence of the various coefficients for Bi₁₂GeO₂₀ on the angle θ between the [001] axis and the direction k of the internal electric field, k being in the (110) plane. (a) Deformation H as defined by Eq. (11). (b) Effective dielectric constant. (c) Ratio r_{eff}/n^2 : solid curve, spatially modulated field; dashed line, uniform field and stress-free sample; dotted line, uniform field and strain-free sample.

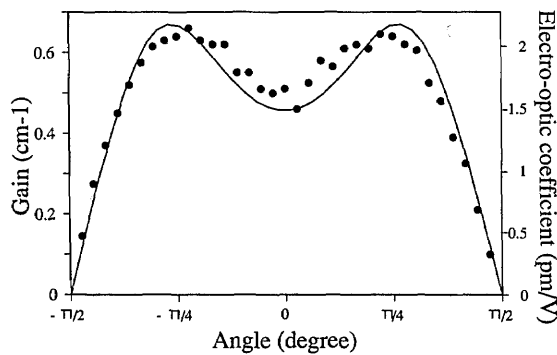


Fig. 3. Experimental data of two-beam-coupling gain (dots) and theoretical plot of the variation (solid curve) of the effective electro-optic coefficient $r_{\text{eff}}^{\text{pr}}/n^4$ versus angle θ , with the same configuration as in Fig. 2 in the case of a modulated field. The sample is a $\text{Bi}_{12}\text{GeO}_{20}$ crystal, and we used the numerical values given in Appendix A for that type of material.

to the effective electro-optic coefficient $r_{\text{eff}}^{\text{pr}}$:

$$r_{\text{eff}}^{\text{pr}} = -v_i^* \frac{\epsilon_0 \Delta \epsilon_{ij}}{E} w_j, \quad (17)$$

which can be rewritten by using Eqs. (3) and (12):

$$r_{\text{eff}}^{\text{pr}} = v_i^* (r_{ijk}^{\delta=0} + p_{ijkl} a_l H) a_k w_j n_i^2 n_j^2. \quad (18)$$

The asterisk denotes complex conjugation.

Equations (15) and (18) show that the constants that are valid for a description of the photorefractive effect are neither the clamped nor the unclamped ones. According to Eq. (8), the crystal must indeed always be considered as internally unclamped along the field axis x' , whatever the applied external forces are. Conversely, the other stress components may be different from zero. Note that the breaking of the crystal symmetry by the spatially modulated field permits a nonzero $r_{\text{eff}}^{\text{pr}}$ coefficient [Eq. (18)] in a configuration (a given field direction and given optical beam polarizations) for which a uniform field [Eq. (5)] would not induce any index change. For example, for a crystal belonging to the point-group symmetry $\bar{4}3m$, a modulated field grating along the [111] crystallographic direction can induce an isotropic coupling between two optical waves polarized along one crystallographic axis. Such an isotropic coupling in these materials is forbidden by the usual symmetry of the electro-optic tensor ($r_{ij} = 0$).

D. Application to BaTiO_3

Experiments are usually performed in BaTiO_3 samples with the induced grating wave vector lying in the (b, c) crystallographic plane. In Fig. 1 we have plotted the dielectric constant $\epsilon_{\text{eff}}^{\text{pr}}$, given by Eq. (15), versus the angle θ between the grating wave vector and the crystallographic b axis. For comparison we have also plotted the effective dielectric constants for a uniform electric field and for totally unclamped (dashed curve) and clamped (dotted curve) crystals. These constants are defined as $\epsilon_{\text{eff}}^{\text{pr}}$ by Eq. (15) with $\epsilon_{ij}^{\sigma=0}$ or $\epsilon_{ij}^{\delta=0}$ substituted for $\epsilon_{ij}^{\text{pr}}$.

Figure 1 shows that the value of the effective dielectric constant that is to be used with a modulated field and in this particular crystallographic orientation lies between the clamped and unclamped values. Unfortunately, we could not find any values for the elasto-optic coefficients

in the literature, and therefore we cannot present any conclusion for the effective electro-optic coefficient.

E. Application to $\text{Bi}_{12}\text{GeO}_{20}$

We consider the special case of a grating with a wave vector lying in the (110) plane, and we study the variation of both $\epsilon_{\text{eff}}^{\text{pr}}$ and $r_{\text{eff}}^{\text{pr}}$ as a function of the angle θ between the [001] axis and the wave vector. In Fig. 2(a) the coefficient H (e.g., the deformation) is plotted versus θ . H shows a maximum for $\theta = \pm 55^\circ$. Figure 2(b) shows the effective dielectric constant for a spatially modulated electric field (solid curve) and for a uniform field for both totally unclamped (dashed line) and clamped (dotted line) samples. In Fig. 2(c) we have plotted the ratio $r_{\text{eff}}^{\text{pr}}/n^4$ (solid curve) seen by two circularly polarized interfering beams with an external incident angle equal to $\pm 15^\circ$. For comparison we have also plotted the effective coefficients by using the totally unclamped and clamped electro-optic tensors in place of the term in parentheses in Eq. (18).

For $\theta = 0^\circ$, $r_{\text{eff}}^{\text{pr}}$ is equal to the clamped value [no deformation as seen in Fig. 2(b)]. However, for $\theta \approx \pm 45^\circ$ $r_{\text{eff}}^{\text{pr}}$ presents two maxima that are larger than both the clamped and unclamped values. For this orientation (electric field along the [111] crystallographic direction) all possible deformations are excited, and they produce the strongest elasto-optic index change. These two maxima were observed by Stepanov *et al.*³ The maxima have equal magnitude because we assume a symmetric elasto-optic tensor: $p_{ijkl} = p_{klij}$. However, that assumption is not necessarily valid for materials that belong to the point-group symmetry 23, so that the two maxima could be of unequal magnitudes.

To check these predictions experimentally, we conducted a two-beam-coupling experiment with a $\text{Bi}_{12}\text{GeO}_{20}$ sample in the configuration described above. Two circularly polarized beams from an Ar^+ laser (514 nm) wrote a grating with a fringe spacing of $1 \mu\text{m}$. The experimental values for the two-wave mixing gain, proportional to $r_{\text{eff}}^{\text{pr}}$, are reported in Fig. 3. The experimental data are in good agreement with the theoretical behavior predicted by Eq. (18).

5. CONCLUSIONS

Piezoelectricity has a large influence on the dielectric and piezoelectric constants in photorefractive materials. In the presence of a uniform electric field these constants depend on externally applied forces. However, those to be used for a spatially modulated field are independent of external forces. Therefore, if one is to describe the photorefractive effect in the presence of an externally applied voltage, different values must be used simultaneously for the dielectric and the electro-optic constants, the first values for the uniform part of the field and the others for its modulated component. We have demonstrated that, because of the breaking of the crystal symmetry by the internal field, the symmetry of the effective electro-optic coefficient is modified in the presence of a photorefractive charge grating. This effective coefficient can be either larger or smaller than both the strain-free (totally clamped) or stress-free (totally unclamped) electro-optic coefficients. The coefficient can also be different from zero in a configuration in which both the clamped and unclamped coefficients vanish.

APPENDIX A

Here nonvanishing coefficients for some photorefractive materials are expressed with abbreviated notations.²

The pairs of indices i, j and k, l (i, j, k , and l from 1 to 3) are contracted to m and n , which take on values from 1 to 6 according to

$$\begin{aligned} 11 \rightarrow 1, \quad 22 \rightarrow 2, \quad 33 \rightarrow 3, \quad 23 \rightarrow 4, \quad 32 \rightarrow 4, \\ 13 \rightarrow 5, \quad 31 \rightarrow 5, \quad 23 \rightarrow 6, \quad 32 \rightarrow 6. \end{aligned}$$

In contrast to tensors \mathbf{s} and \mathbf{d} , for those components given below no multiplying factor must be introduced between the full and abbreviated notations, so that for any i, j, k , or l we always have

$$\begin{aligned} c_{ijkl} = c_{mnn}, \quad \epsilon_{ij} = \epsilon_m, \quad e_{ikl} = e_{im}, \\ r_{ijk} = r_{mk}, \quad p_{ijkl} = p_{mn}. \end{aligned}$$

BaTiO₃, Point-Group Symmetry 4mm

Rigidity tensor⁷ $c_{ij} = c_{ji}$ (10^{10} N/m²):

$$\begin{aligned} c_{11} = c_{22} = 24.5, \quad c_{12} = 12.8, \quad c_{13} = 12.3, \\ c_{33} = 14.8, \quad c_{44} = c_{55} = 5.5, \quad c_{66} = 12.0. \end{aligned}$$

Stress-free dielectric constant⁷:

$$\epsilon_1 = \epsilon_2 = 4000, \quad \epsilon_3 = 130.$$

Piezoelectric tensor⁷ (C/m²):

$$e_{15} = e_{24} = 35.6, \quad e_{31} = e_{32} = -3.96, \quad e_{33} = 1.92.$$

Stress-free electro-optic tensor^{8,9} (pm/V):

$$r_{13} = r_{23} = 19.5, \quad r_{33} = 97, \quad r_{42} = 1640.$$

GaAs, Point-Group Symmetry $\bar{4}3m$

Rigidity tensor¹⁰ $c_{ij} = c_{ji}$ (10^{10} N/m²):

$$\begin{aligned} c_{11} = c_{22} = c_{33} = 11.90, \quad c_{12} = c_{13} = c_{23} = 5.38, \\ c_{44} = c_{55} = c_{66} = 5.95. \end{aligned}$$

Stress-free dielectric¹⁰ constant at $T = 300$ K:

$$\epsilon_1 = \epsilon_2 = \epsilon_3 = 12.85.$$

Piezoelectric tensor¹⁰ (C/m²):

$$e_{14} = e_{15} = e_{36} = 0.154.$$

Stress-free electro-optic tensor¹¹ (pm/V):

$$r_{41} = r_{52} = r_{63} = 1.2.$$

Elasto-optic tensor¹² ($p_{ij} = p_{ji}$):

$$\begin{aligned} p_{11} = p_{22} = p_{33} = 0.165, \quad p_{12} = p_{13} = p_{23} = 0.14, \\ p_{44} = p_{55} = p_{66} = 0.072. \end{aligned}$$

Bi₁₂GeO₂₀, Point-Group Symmetry 23

Rigidity tensor¹³ $c_{ij} = c_{ji}$ (10^{10} N/m²):

$$\begin{aligned} c_{11} = c_{22} = c_{33} = 12.8, \quad c_{12} = c_{13} = c_{23} = 3.05, \\ c_{44} = c_{55} = c_{66} = 2.55. \end{aligned}$$

Stress-free dielectric constant¹⁴:

$$\epsilon_1 = \epsilon_2 = \epsilon_3 = 48.$$

Piezoelectric tensor¹³ (C/m²):

$$e_{14} = e_{15} = e_{36} = 0.98.$$

Stress-free electro-optic tensor¹⁵ (pm/V):

$$r_{41} = r_{52} = r_{63} = 3.6.$$

Elasto-optic tensor³ ($p_{ij} \neq p_{ji}$):

$$\begin{aligned} p_{11} = p_{22} = p_{33} = 0.12, \quad p_{12} = p_{13} = p_{23}, \\ p_{21} = p_{31} = p_{32}, \end{aligned}$$

with

$$p_{12} + p_{13} = 0.19, \quad p_{44} = p_{55} = p_{66} = 0.01.$$

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