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Relations between electrostriction and the stress-optical effect*

Albert Feldman
National Bureau of Standards, Washington, D. C. 20234
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Relationships are obtained between the stress-optical constants and the electrostrictive coefficients of solid materials. These results, which are dependent upon the geometric boundary conditions, encompass the apparent differences in the relationships derived by different authors.

The process of electrostriction is of current interest because it can cause the self-focusing of high-intensity radiation in solids. Articles have been published that relate the electrostrictive coefficients to the stressoptic,[1] elasto-optic,[2] or piezodielectric coefficients[3] with differing results. In an early work, Guggenheim[4] has derived similar relations for liquids which undergo magnetostriction. He showed that the particular relationship obtained will depend upon the boundary conditions much as the electric polarization in a solid depends upon the shape of the solid. Guggenheim showed that solutions are easily obtained only for relatively simple configurations.

In this paper, we derive relationships between the electrostrictive coefficients and the stress-optical constants for dielectric materials with inversion symmetry, based on Guggenheim's work. We ignore any effects due to body rotations. The three cases treated are shown in Fig. 1. Each case, which corresponds to a different set of boundary conditions, results in a different relationship between the photoelastic constants and the electrostrictive coefficients. Thus, the electrostrictive strain induced in a solid located in an external electric field will depend upon the shape of the solid and the orientation of the solid in the field.

The following notation is used in the analysis: V_E is the volume occupied by the electric field; V_S is the volume of the solid in the absence of strain; κ_{ij} is an element of the dielectric tensor and, κ_{ij}^{-1} is an element of its inverse tensor; ϵ_{ij} is an element of the strain tensor; σ_{ij} is an element of the mechanical stress tensor; q_{ijkl} is a stress-optical constant; p_{ijkl} is an elasto-optic coefficient; γ_{ijkl} is an electrostriction coefficient; and s_{ijkl} is an elastic compliance coefficient. The phenomenological relationships among these constants are

$$D_i=\kappa_{ij}E_j\,,\quad E_i=\kappa_{ij}^{-1}D_j\,, \tag{1}$$

$$\Delta\kappa_{ij}^{-1}=q_{ijkl}\sigma_{kl}=p_{ijkl}\epsilon_{kl}\,, \tag{2}$$

$$\epsilon_{kl}=\tfrac{1}{2}\gamma_{ijkl}E_iE_j+s_{klmn}\sigma_{mn}\,. \tag{3}$$

In Eq. (2), we have ignored the term quadratic in electric field, the Kerr effect term. Equations (1) and (2) are taken to apply when the fields are either constant or time-varying and stresses and strains are constant in time. When $\vec{E}$ is varying rapidly in time so that $\underline{\epsilon}$ cannot respond to the instantaneous value of $\vec{E}$, we take a time average of E_iE_j in Eq. (3). The tensors $\underline{\kappa}$, $\underline{\kappa}^{-1}$, $\underline{q}$, $\underline{p}$, and $\underline{\gamma}$ will then depend upon the frequency of the field.

Following Guggenheim, we can write the free energy of a system in one of the following forms, depending on which variables are meant to be independent:

$$F_1=U-TS-\int\sigma_{ij}\epsilon_{ij}\,dV-\frac{1}{4\pi}\int E_iD_i\,dV\quad(T,\vec{E},\underline{\sigma}), \tag{4a}$$

$$F_2=U-TS-\int\sigma_{ij}\epsilon_{ij}\,dV\quad(T,\vec{D},\underline{\sigma}), \tag{4b}$$

where, in the convention used, the independent variables are listed to the right. F_1 is the Gibbs function and F_2 is the elastic Gibbs function.[5] We integrate over a fixed volume of space, which includes V_E and V_S; U is the internal energy; T is the temperature; S is the entropy. The change in internal energy of the system is given by[6]

$$\delta U=TdS+\frac{1}{4\pi}\int E_i\delta D_i\,dV+\int\sigma_{ij}\delta\epsilon_{ij}\,dV\quad(S,\vec{D},\underline{\epsilon}).$$

We then obtain

$$\delta F_1=-SdT-\frac{1}{4\pi}\int D_i\delta E_i\,dV-\int\epsilon_{ij}\delta\sigma_{ij}\,dV\quad(T,\vec{E},\underline{\sigma}), \tag{5a}$$

$$\delta F_2=-SdT+\frac{1}{4\pi}\int E_i\delta D_i\,dV-\int\epsilon_{ij}\delta\sigma_{ij}\,dV\quad(T,\vec{D},\underline{\sigma}). \tag{5b}$$

Similar expressions can be derived with S and $\underline{\epsilon}$ as independent variables. The above expressions will be used to obtain the desired relationships between $\underline{\gamma}$, $\underline{q}$, and $\underline{\kappa}$.

Case I. We have a solid of volume V_S with a uniform electric field totally enclosed within V_S, except for in a thin layer of material at the boundary of the solid in which the field falls to zero. When the solid undergoes a strain, we assume that the electric field is still enclosed within the solid, but V_E is unchanged. We choose this geometry in order to neglect the stresses developed because of the constraining effect of the field-free region of the solid. For sufficiently large volumes, V_S can be

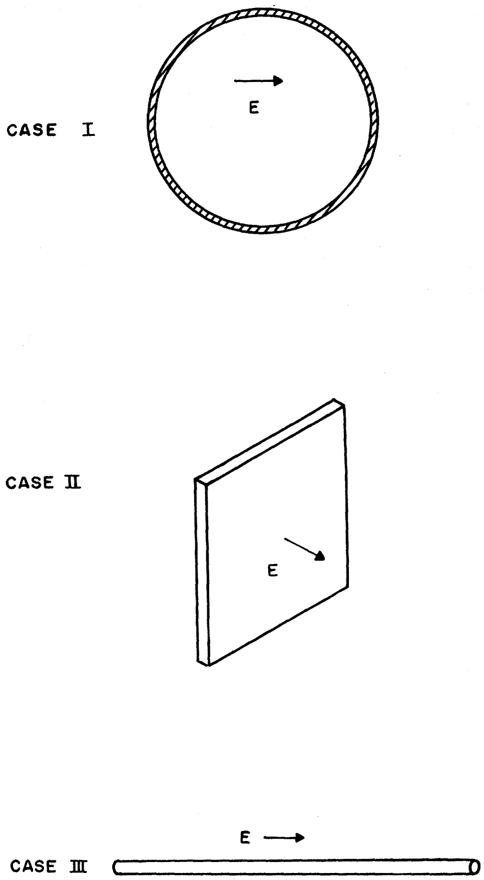

FIG. 1. Configurations for which relationships are derived between the electrostrictive coefficients and the stress-optical constants: Case I—a uniform electric field confined within a solid; case II—a thin slab of material in a uniform external electric field with the large slab faces perpendicular to the field; case III—a long narrow cylinder whose axis lies parallel to a uniform external electric field.

considered equal to V_E. Using Eq. (5a), we obtain

$$dF_1 = -SdT - (V_E/4\pi)\kappa_{ij}E_i dE_j - V_S\epsilon_{ij}d\sigma_{ij}. \qquad (6)$$

For an isothermal process

$$\kappa_{ij} = -\frac{4\pi}{V_S}\left(\frac{\partial^2 F_1}{\partial E_i \partial E_j}\right)_{T,\underline{\sigma}} \qquad (7)$$

and

$$\epsilon_{ij} = -\frac{1}{V_S}\left(\frac{\partial F_1}{\partial \sigma_{ij}}\right)_{T,\vec{E}}. \qquad (8)$$

Taking higher derivatives, we obtain

$$(1/4\pi)\partial\kappa_{ij}/\partial\sigma_{kl} = \partial^2\epsilon_{kl}/\partial E_i \partial E_j. \qquad (9a)$$

Using Eq. (5b), we obtain a similar result

$$\frac{1}{4\pi}\frac{\partial\kappa^{-1}_{ij}}{\partial\sigma_{kl}} = -\frac{\partial^2\epsilon_{kl}}{\partial D_i \partial D_j}. \qquad (9b)$$

Equations (1), (3), and (9a) or (9b) lead to the equality

$$\gamma_{ijkl} = -(1/4\pi)\kappa_{im}\kappa_{jn}q_{mnkl}. \qquad (10)$$

Equation (10) has been used for calculating the electrostrictive strains induced by focusing a laser beam into the interior of a solid.[7,8] It corresponds to the results of Refs. 1 and 3.

Case II. Consider a thin slab of material in a uniform external $\vec{E}$ field with the large faces of the slab perpendicular to the field. In this case it is convenient to use Eq. (5b) because $\vec{D}$ is continuous across the slab boundary. If end effects are neglected, we obtain

$$dF_2 = -SdT + (V_E/4\pi)D_i dD_i + (1/4\pi)(\kappa^{-1}_{ij} - \delta_{ij}) \times (1+\epsilon_{mn}\delta_{mn})V_S D_i dD_j - V_S\epsilon_{ij}d\sigma_{ij}. \qquad (11)$$

The second and third terms of this equation account for the electrical energy of the vacuum plus the electrical energy of the strained solid. Taking derivatives for the isothermal case, we find

$$\epsilon_{kl} = -\frac{1}{V_S}\left(\frac{\partial F_2}{\partial\sigma_{kl}}\right)_{T,\vec{D}}; \qquad (12)$$

$$\left(\frac{\partial^2 F_2}{\partial D_i \partial D_j}\right)_{T,\underline{\sigma}} = \frac{V_S}{4\pi}(\kappa^{-1}_{ij} - \delta_{ij})(1+\epsilon_{mn}\delta_{mn}) + \frac{V_E}{4\pi}\delta_{ij}. \qquad (13)$$

Higher-order derivatives lead to the result

$$\frac{\partial^2\epsilon_{kl}}{\partial D_i \partial D_j} = -\frac{1}{4\pi}\frac{\partial\kappa^{-1}_{ij}}{\partial\sigma_{kl}} - \frac{1}{4\pi}(\kappa^{-1}_{ij} - \delta_{ij})\delta_{mn}s_{mnkl} \qquad (14)$$

or

$$\gamma_{pqkl} = -(1/4\pi)\kappa_{pi}\kappa_{qj}[q_{ijkl} + (\kappa^{-1}_{ij} - \delta_{ij})\delta_{mn}s_{mnkl}], \qquad (15)$$

where we have kept terms to lowest order.

Case III. Consider a long narrow cylinder whose axis lines up parallel to a uniform electric field. In this case it is convenient to use Eq. (5a) because $\vec{E}$ is continuous across the cylinder wall. Neglecting end effects we obtain

$$dF_1 = -SdT - (V_E/4\pi)E_i dE_i - (1/4\pi)(\kappa_{ij} - \delta_{ij}) \times (1+\epsilon_{mn}\delta_{mn})V_S E_i dE_j - V_S\epsilon_{ij}d\sigma_{ij}. \qquad (16)$$

Taking derivatives for the isothermal case, following the procedures of cases I and II and keeping terms of lowest order, we obtain

$$\frac{\partial^2\epsilon_{kl}}{\partial E_i \partial E_j} = \frac{1}{4\pi}\frac{\partial\kappa_{ij}}{\partial\sigma_{kl}} + \frac{1}{4\pi}(\kappa_{ij} - \delta_{ij})\delta_{mn}s_{mnkl}. \qquad (17)$$

But, the susceptibility is $\chi_{ij} = (\kappa_{ij} - \delta_{ij})/4\pi$, so that

$$\gamma_{ijkl} = -(1/4\pi)\kappa_{im}\kappa_{jn}q_{mnkl} + \chi_{ij}\delta_{mn}s_{mnkl}. \qquad (18)$$

Equation (18) corresponds to the results of Maradudin and Burstein.[2]

The terms which occur in case II [Eq. (15)] and case III [Eq. (18)] but are absent in case I [Eq. (10)] are due to the interchange of solid dielectric properties with vacuum dielectric properties in the incremental volume at the boundary of the solid, produced by the strain. The difference between these terms results from the angle the electric field makes with the solid boundary. This angle affects the electrical energy density within the solid at the boundary and hence affects the free energy of the system.

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