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ABSTRACT

Theories of electro- and magneto-effects are reviewed. Symmetry argument for finding independent and nonvanishing components of the electro- and magneto-optical tensor coefficients is briefly described. Emphasis is on the recent development of the microscopic theories which predict quite accurately the magnitudes of these nonvanishing components.

I. INTRODUCTION

The properties of a material may vary with applied electric and magnetic fields. In particular, the fields induce a change in the optical dielectric constants ε_{ij} or the refractive indices n_{ij}. Consequently, light propagation in the material is affected by the fields. Such induced optical effects are known as electro-optical[1] (induced by electric field only) and magneto-optical[2] (induced by magnetic field only) effects. Thus, for examples, a dc electric field can induce linear birefringence and a dc magnetic field can induce both linear and circular birefringences in a medium. Electro- and magneto-optical effects are the underlying principles for many useful optical devices. Scientifically, measurements of electro- and magneto-optical effects often yield valuable information about the structure and electronic properties of a material.

In the presence of the applied dc fields $\underset{\sim}{E}^{dc}$ and $\underset{\sim}{H}^{dc}$, the optical dielectric constants of a medium can be written as

$$\varepsilon_{ij}(\underset{\sim}{E}^{dc}, \underset{\sim}{H}^{dc}) = \varepsilon^{o}_{ij} + \Delta\varepsilon_{ij}(\underset{\sim}{E}^{dc}, \underset{\sim}{H}^{dc}). \tag{1}$$

Two questions arise in the discussion of electro- and magneto-optical effects: (1) For a given medium, what do we know about the tensor $\underset{\approx}{\Delta\varepsilon}$? (2) Given $\underset{\approx}{\Delta\varepsilon}$, how does it affect light propagation in the medium? In principle, the second question can always be answered by solving the Maxwell equations. Here, we simply refer to the literatures on the subject.[3] In this review, we shall limit ourselves to the discussion of the first question.

The dielectric constants of Eq. (1) can usually be expanded into power series of $\underset{\sim}{E}^{dc}$ and $\underset{\sim}{H}^{dc}$.

$$\varepsilon_{ij} = \varepsilon^{o}_{ij} + 4\pi \Big[\chi^{(2e)}_{ijk}\, E^{dc}_{k} + \chi^{(2b)}_{ijk}\, H^{dc}_{k}$$

$$+ \chi^{(3ee)}_{ijk\ell}\, E^{dc}_{k} E^{dc}_{\ell} + \chi^{(3eb)}_{ijk\ell}\, E^{dc}_{k} H^{dc}_{\ell} + \chi^{(3bb)}_{ijk\ell}\, H^{dc}_{k} H^{dc}_{\ell} + \ldots \Big] \qquad (2)$$

where χ's are coefficients independent of the fields,[4] and the convention that a tensor product is expressed by repeated subindices is used. For simplicity, we shall assume in this paper that ε_{ij} depends weakly on the applied fields, and therefore only the leading terms in Eq. (2) give observable effects. In some special cases, one can also include the full expression of ε_{ij} in the description without too much complication.

Our problem now is to find $\underset{\approx}{\chi}$'s in Eq. (2) for a given medium. Because of the existing symmetry of the medium, many components of the $\underset{\approx}{\chi}$ tensors are vanishing and only a few independent. The problem can then be separated into two parts: first, to find from the existing symmetry of a given medium the independent nonvanishing components of the $\underset{\approx}{\chi}$ tensors and then, to find the magnitudes of these nonvanishing components. While the second part of the problem depends on the microscopic properties of the medium, the first part can be solved by a simple thermodynamic treatment as we shall now briefly describe.

Again, for simplicity, we shall assume that the medium is nondissipative. Optical effects at absorbing frequencies are, of course, important, but they yield more specific information about the energy states involved in the absorption process than general information about the medium as a whole. Let us consider a quasi-monochromatic light which is represented by the field

$$\underset{\sim}{E}(t) = \underset{\sim}{\mathcal{E}}(t) \exp(-i\omega t) + \underset{\sim}{\mathcal{E}}^{*}(t) \exp(i\omega t) \tag{3}$$

where $|\partial\underset{\sim}{\mathcal{E}}/dt| << |\omega\underset{\sim}{\mathcal{E}}|$. If the light is propagating in a nondissipative medium with a dielectric constant $\underset{\approx}{\varepsilon}(\omega)$, then the time-averaged energy density stored in the medium is given by[5]

$$\bar{U} = (1/4\pi) \left[\mathcal{E}_i^{*} (\partial\omega\varepsilon_{ij}/\partial\omega)\, \mathcal{E}_j + |H|^2\right] \tag{4}$$

which becomes, with the expression of ε_{ij} in Eq. (2)

$$\begin{aligned} \bar{U} = (1/4\pi)\Big\{&\mathcal{E}_i^{*} (\partial\omega\varepsilon^{o}_{ij}/\partial\omega)\, \mathcal{E}_j + |H|^2 \\ &+ 4\pi\, \mathcal{E}_i^{*} (\partial/\partial\omega)\omega\Big[\chi^{(2e)}_{ijk} E^{dc}_k + \chi^{(2b)}_{ijk} H^{dc}_k \\ &+ \chi^{(3ee)}_{ijk\ell} E^{dc}_k E^{dc}_\ell + \chi^{(3eb)}_{ijk\ell} E^{dc}_k H^{dc}_\ell + \chi^{(3bb)}_{ijk\ell} H^{dc}_k H^{dc}_\ell + \ldots\Big] \mathcal{E}_j\Big\}. \end{aligned} \tag{5}$$

Suppose we now rotate the medium about a certain axis by a certain angle. This energy density should change accordingly from $\bar{U}$ to $\bar{U}'$. If, however, the rotation happens to be a symmetry operation in the symmetry group of the medium, then the medium would appear unchanged after the rotation, and therefore $\bar{U} = \bar{U}'$. This symmetry argument applies to each term of Eq. (5) independently, and can be used to find the independent components of $\underset{\approx}{\chi}$.

Let us consider as an example the $\chi^{(2e)}$ term which is responsible for the linear electro-optical effect. We write

$$\mho_e = \mathcal{E}_i^* \,(\partial\omega\chi_{ijk}^{(2e)}/\partial\omega)\, \mathcal{E}_j E_k^{dc}. \tag{6}$$

If S_{ij} represents a symmetry operation, then we would have

$$\mho'_e = \mathcal{E}^*_{i'},\, S^*_{ii'}\, (\partial\omega\chi^{(2e)}_{ijk}/\partial\omega)\, S_{jj'},\, S_{kk'},\, E_{j'},\, E^{dc}_{k'} = \mho_e \tag{7}$$

and hence

$$S^*_{ii'},\, \chi^{(2e)}_{ijk}\, S_{jj'},\, S_{kk'} = \chi^{(2e)}_{i'j'k'} \tag{8}$$

we have n such relations if there are n symmetry operations in the symmetry group of the medium. In addition, we have, from general principle of symmetry for a nondissipative medium,

$$\varepsilon_{ij} = \varepsilon^*_{ji}. \tag{9}$$

These relations make some components of $\underset{\approx}{\chi}(2e)$ vanish and others depend on one another. An immediate consequence of Eq. (7) or (8) is that all components of $\underset{\approx}{\chi}(2e)$ should be zero for crystals with inversion symmetry. As a simple illustration, let us consider the zincblende crystals, such as GaAs, which belong to the class T_d. These crystals remain invariant under 180° rotations about the three 4-fold axes. From Eq. (7) or (8), we find

$$\begin{gathered} \chi^{(2e)}_{iii} = -\chi^{(2e)}_{iii} = 0; \\ \chi^{(2e)}_{ijj} = -\chi^{(2e)}_{ijj} = 0;\ \chi^{(2e)}_{iij} = -\chi^{(2e)}_{iij} = 0,\ i \neq j; \end{gathered} \tag{10}$$

and $\chi^{(2e)}_{ijk}$ with $i \neq j \neq k$ are the only nonvanishing components. The crystals also remain invariant under mirror reflections about the three diagonal planes. This symmetry gives

$$\begin{aligned} \chi^{(2e)}_{xyz} &= \chi^{(2e)}_{yzx} = \chi^{(2e)}_{zxy}, \\ &= \chi^{(2e)}_{yxz} = \chi^{(2e)}_{zyx} = \chi^{(2e)}_{xzy} \end{aligned} \tag{11}$$

and, consequently, we have only one independent component of $\underset{\approx}{\chi}^{(2e)}$ for crystals in the class T_d. It can be seen that the linear electro-optical effect and the piezoelectric effect follow the same symmetry considerations. We can, therefore, obtain the linear electric-optical tensor $\underset{\approx}{\chi}^{(2e)}$ for a crystal from the piezoelectric tensor listed in many texts.[6]

The quadratic electro-optical tensor $\underset{\approx}{\chi}^{(3ee)}$ for a medium can be determined similarly. For example, for an isotropic medium, the nonvanishing components of $\underset{\approx}{\chi}^{(3ee)}$ are $\chi^{(3ee)}_{iiii}$, and $\chi^{(3ee)}_{iijj}$ with $i \neq j$.

The symmetry considerations for the terms involving magnetic field in Eq. (5) are slightly different, since the magnetic field, being a pseudovector, is invariant under inversion. For a pseudovector, the transformation matrix for a pure rotation is the same as that for a rotation plus an inversion. Therefore, we should have

$$S^{*}_{ii'}\, \chi^{(2b)}_{ijk}\, S_{jj'}\, S^{M}_{kk'} = \chi^{(2b)}_{i'j'k'}, \text{ etc.} \tag{12}$$

where $S^{M}_{kk'}$ are transformation matrices for symmetry operations associated with a pseudovector. As a consequence $\underset{\approx}{\chi}^{(2b)}$, unlike $\underset{\approx}{\chi}^{(2e)}$, does not vanish in a medium with inversion symmetry. For example, in an isotropic medium or a cubic system, the nonvanishing components of $\underset{\approx}{\chi}^{(2b)}$ are $\chi^{(2b)}_{xyz} = \chi^{(2b)}_{yzx} = \chi^{(2b)}_{zxy} = \chi^{(2b)*}_{yxz} = \chi^{(2b)*}_{zyx} = \chi^{(2b)*}_{xzy}$. In addition to Eqs. (9) and (12), one can also show from general principle of symmetry that[7]

$$\varepsilon_{ij}\,(\underset{\sim}{H}^{dc}) = \varepsilon_{ji}\,(-\underset{\sim}{H}^{dc})\ . \tag{13}$$

As a result, the real part of ε_{ij} should be an even function of $\underset{\sim}{H}^{dc}$ and the imaginary part of ε_{ij} an odd function of $\underset{\sim}{H}^{dc}$. Physically, the imaginary part of ε_{ij} induced by the magnetic field leads to induced circular birefringence or the Faraday effect,[2] and the real part leads to induced linear birefringence or the Voigt effect.[8] We can conclude from Eqs. (9) and (13) that all the nonvanishing components of $\chi_{ijk}^{(2b)}$ and $\chi_{ijk\ell}^{(3eb)}$ are imaginary and those of $\chi_{ijk}^{(2e)}$, $\chi_{ijk\ell}^{(3ee)}$, and $\chi_{ijk\ell}^{(3bb)}$ real.

We note in passing that the same energy density $\bar{U}$ of Eq. (5) also governs the inverse electro- and magneto-optical effects. If we consider $\bar{U}$ as the stored energy density of the dc fields, then the corresponding dc polarization $\underset{\sim}{P}^{dc}$ and magnetization $\underset{\sim}{M}^{dc}$ are[9]

$$P_k^{dc} = \mathcal{E}_i^* \, (\partial/\partial\omega)\omega\left[\chi_{ijk}^{(2e)} + \chi_{ijk\ell}^{(3eb)} H_\ell^{dc} + 2\chi_{ijk\ell}^{(3ee)} E_\ell^{dc}\right]\mathcal{E}_j \tag{14}$$

$$M_k^{dc} = \mathcal{E}_i \, (\partial/\partial\omega)\omega\left[\chi_{ijk}^{(2b)} + \chi_{ijk\ell}^{(3eb)} E_\ell^{dc} + 2\chi_{ijk\ell}^{(3bb)} H_\ell^{dc}\right]\mathcal{E}_j . \tag{15}$$

Both $\underset{\sim}{P}^{dc}$ and $\underset{\sim}{M}^{dc}$ are quadratic functions of the optical field. These equations show that dc polarization and magnetization can be induced by an optical field. The inverse electro-optical effect, known as optical rectification, has been observed in electro-optical crystals ($\underset{\approx}{\chi}^{(2e)} \neq 0$).[10] The inverse Faraday effect, which comes from $\underset{\approx}{\chi}^{(2b)}$, has also been observed in both paramagnetic and diamagnetic materials.[11] Calculations show that the inverse Voigt effect, arising from the $\underset{\approx}{\chi}^{(3bb)}$ term, should also be observable.[12,13]

We have seen how symmetry considerations can help us determine the forms of the $\underset{\approx}{\chi}$ tensors for a given medium. We now come to the central theme of this review--how to find the magnitudes of these $\underset{\approx}{\chi}$ coefficients.[14] In the following sections, we shall discuss electro-optical and magneto-optical effects separately. We shall first outline the procedure for deriving the exact microscopic expressions for $\underset{\approx}{\chi}$. We shall then use a two-level or two-band model to simplify the expressions, so that quantitative estimates of χ become feasible.

II. LINEAR ELECTRO-OPTICAL EFFECT

In general, both electrons and ions in a medium contribute to electro-optical effects. At optical frequencies much larger than the frequencies of lattice vibration, the ionic contribution comes in because the dc electric field disturbs the lattice, but the effect is often negligible, or can be taken into account implicitly through its effect on dc susceptibility. Here, we shall limit our discussion to the electronic contribution only.

The microscopic expression for $\chi_{ijk}^{(2e)}$ can be obtained from straight forward perturbation calculation. In the electric-dipole approximation, the Hamiltonian of the system is given by

$$\mathcal{H} = \mathcal{H}_o - e\underset{\sim}{r}\cdot(\underset{\sim}{E}^{dc} + \underset{\sim}{\mathcal{E}}\, e^{-i\omega t} + \underset{\sim}{\mathcal{E}}^{*}\, e^{i\omega t}) \qquad (16)$$

where $\mathcal{H}_o$ is the Hamiltonian for the unperturbed material system, $\underset{\sim}{r}$ is the electron coordinate with respect to the position of the associated molecule or the unit cell, and the symbol for summation over electrons is omitted for simplicity. The first-order perturbation theory with $\underset{\sim}{E}^{dc} = 0$ leads to the usual expression for the linear dielectric constant[15]

$$\epsilon_{ij} = 1 + 4\pi\,\chi_{ij}$$

$$\chi_{ij} = Ne^2 \sum_{n,n'} \left\{ \frac{(r_i)_{nn'}\,(r_j)_{n'n}}{\hbar(\omega_{n'n} - \omega)} + \frac{(r_j)_{nn'}\,(r_i)_{n'n}}{\hbar(\omega_{n'n} + \omega)} \right\} \rho_n^o \qquad (17)$$

where

$$(r_i)_{nn'} = \langle n|r_i|n'\rangle$$

$$\mathcal{H}_o|n\rangle = E_n|n\rangle$$

$$\hbar\omega_{n'n} = E_{n'} - E_n ,$$

N is the density of molecules[16] or unit cells, and ρ_n^o is the population in the $|n\rangle$ state at thermal equilibrium. Application of a dc electric field on the system modifies the linear susceptibility χ_{ij} through changes in the energies, the wave functions, and the populations of the electronic states. In most electro-optical materials, the states are either fully occupied or empty at normal temperature, and the change in ρ_n^o is negligible. Then, the next-order perturbation calculation yields the microscopic expression for $\chi_{ikj}^{(2e)}$ [17-19]

$$\chi_{ijk}^{(2e)} = (Ne^3/\hbar^2) \sum_{g,n,n'} \left\{ \frac{\omega_{ng}\,(r_i)_{gn}\,(r_k)_{nn'}\,(r_j)_{n'g}}{(\omega^2 - \omega_{ng}^2)\,\omega_{n'g}} + \frac{(\omega^2 + \omega_{ng}\,\omega_{n'g})(r_i)_{gn}(r_j)_{nn'}(r_k)_{n'g}}{(\omega^2 - \omega_{ng}^2)(\omega^2 - \omega_{n'g}^2)} + \frac{\omega_{n'g}\,(r_j)_{gn}\,(r_i)_{nn'}\,(r_k)_{n'g}}{(\omega^2 - \omega_{n'g}^2)\,\omega_{ng}} \right\} \tag{18}$$

where $|g\rangle$ are the fully occupied ground states and $|n\rangle$ and $|n'\rangle$ are the empty excited states.

It is obvious from the above expression that without detailed knowledge about energies and wave functions of all the electronics states, one cannot possibly evaluate $\chi_{ijk}^{(2e)}$. Unfortunately, our knowledge about excited electronic states is very limited. To find the magnitudes of $\chi^{(2e)}_{ijk}$, we must resort to some approximation to circumvent this difficulty. The usual approximation is to assume an effective mean transition frequency $\bar{\omega}_{ng}$ for all transitions. It is equivalent to approximating the complex system by a simple two-level system. The approximation is reasonable if the integrated oscillator strength for transitions in a fairly narrow range of frequencies dominates, and if the frequency ω is far away from any resonance absorption. By pulling the dispersion factors outside the summation sign, we can then write[19, 20]

$$\sum_{n,n'} (r_i)_{gn} (r_j)_{nn'} (r_k)_{n'g} = (r_i r_j r_k)_{gg} \qquad (19)$$

assuming $(r)_{gg} = 0$. Consequently, Eq. (18) reduces to the form[19]

$$\chi^{(2e)}_{ijk} = (Ne^3/\hbar^2) \, [(3\omega^2 - \bar{\omega}^2_{ng})/(\omega^2 - \bar{\omega}^2_{ng})^2] \sum_g (r_i r_j r_k)_{gg}. \qquad (20)$$

Then, for evaluating $\chi^{(2e)}_{ikj}$, we only need to know $\bar{\omega}_{ng}$ and the wave functions for the ground states $|g\rangle$.

To find $\bar{\omega}_{ng}$, we assume that the same mean frequency can be used to reduce χ_{ii}, so that Eq. (17) gives

$$\chi_{ii} = [2Ne^2\bar{\omega}_{ng}/\hbar(\bar{\omega}^2_{ng} - \omega^2)] \sum_g (r_i r_i)_{gg}. \qquad (21)$$

The above expression describes reasonably well the dispersion of the linear susceptibility for many materials. We can therefore deduce $\bar{\omega}_{ng}$ from measurements of refractive indices at several frequencies.

The ground-state wave functions or electron distribution depend critically on the microscopic structure of the medium. However, in most cases, they can be approximated by simple linear combinations of atomic orbitals or other basic functions such as the Bloch functions. Thus, for example, in a III - V compound, the valence electrons are mainly responsible for the optical transitions. The valence states therefore correspond to the ground states $|g\rangle$ in Eqs. (20) and (21). The molecular model of a III - V compound ($A_{III}B_V$) assumes that the eight valence electrons around each A_{III} atom are paired up to form four tetrahedral bonds connecting the A_{III} atom to the four adjacent B_V atoms as shown in Fig. 1. From the theory of molecular orbitals,[21] we can then describe the ground-state wave functions of these valence electrons approximately by the linear combinations of sp^3 hybridized orbitals centered at A_{III} and B_V and pointing in the [1,1,1] directions.

$$\begin{aligned} \psi_{III-V} &= (\lambda\psi_{III} + \psi_V)/(1 + \lambda^2) \\ \psi_{III} &= \frac{1}{2}(\phi_{ns} + \sqrt{3}\,\phi_{np_\xi}) \\ \psi_V &= \frac{1}{2}(\phi_{n's} - \sqrt{3}\,\phi_{n'p_\xi}) \end{aligned} \tag{22}$$

where ϕ_{ns}, ϕ_{np_ξ}, etc. are hydrogen-like atomic wave functions, ψ_{III} describes an orbital centered at A_{III} and pointing in the bond direction $\hat{\xi}$, ψ_V is centered at B_V and pointing in the $-\hat{\xi}$ direction, and λ is a parameter which determines the ionicity or polarity of the bond.[22] Thus, knowing the ground-state wave functions, we can readily evaluate $\chi^{(2e)}_{ijk}$ from Eq. (20) by summing over contributions from the eight valence electrons per each A_{III} atom.[20,23] The factor N in Eq. (20) is now the density of A_{III} atoms, and the overlapping of bond orbitals has been neglected.

A somewhat different approach has been taken by Levine[24] using Phillips' theory of chemical bond. Again assume that we can replace all the transition frequencies in a semiconductor or insulator by the mean frequency $\bar{\omega}_{ng}$. Then, the Thomas-Reiche-Kuhn sum rule becomes[25]

$$\sum_n \frac{2m}{\hbar} \omega_{ng} \left|(r_i)_{ng}\right|^2 = \frac{2m}{\hbar} \bar{\omega}_{ng} (r_i r_i)_{gg} = 1. \tag{23}$$

Along a crystal axis, Eq. (21) reduces to the form

$$\chi_{ii} = \hbar^2\omega_p^2/(\bar{E}_g{}^2 - \hbar^2\omega^2) \tag{24}$$

where $\omega_p^2 = \mathcal{N}e^2/m$ is the plasma frequency with $\mathcal{N}$ being the density of valence electrons, and $\bar{E}_g = \hbar\bar{\omega}_{ng}$ is the effective energy gap, which is usually larger than the true energy gap. In the limit of zero frequency, we obtain $\chi_{ii} = \hbar^2\omega_p^2/\bar{E}_g^2$, which has been derived by Penn from band-theory calculation.[26] Application of a dc electric field should modify χ_{ii} through $\bar{E}_g$. We can express χ_{ii} in terms of the bond polarizability $\alpha_{\xi\xi}$ along the bond direction $\hat{\xi}$, assuming that the bond polarizability in the direction perpendicular to $\hat{\xi}$ is negligible,

$$\chi_{ii} = \sum_\xi T_{i\xi} \mathcal{N}_\xi \alpha_{\xi\xi} \tag{25}$$

where $T_{i\xi}$ is a geometric factor arising from coordinate transformation between $\hat{\xi}$ and $\hat{i}$, and $\mathcal{N}_\xi$ is the density of bonds along $\hat{\xi}$. If all bonds are alike except pointing in different directions, then we have

$$\chi_{ii} = F_{ii}(\hat{\xi})\, \chi_{\xi\xi} \tag{25'}$$

with $F_{ii}(\hat{\xi}) = \sum_\xi T_{i\xi}{}^2 \mathcal{N}_\xi/\mathcal{N}$. This is true in many simple crystals. From a two-band model, Phillips has shown that[27]

$$\bar{E}_g = [E_h^2 + C_\xi^2]^{(1/2)}$$

$$C_\xi = f\ e^2[(Z_A/r_A) - (Z_B/r_B)] \tag{26}$$

where E_h is the homopolar effective energy gap determined mainly by the lattice constant, C_ξ is the charge-transfer energy, Z_A and r_A are respectively the number of valence electrons and the covalent radius of the A atom in the ξ bond, and f represents the effects of Thomas-Fermi screening ($f \approx 0.12$).[24] From Eq. (26), one can now calculate $\bar{E}_g$ numerically for a given simple crystal.

We are, however, interested in finding $\underset{\approx}{\chi}^{(2e)}$. As Levine has pointed out,[24] an applied dc electric field should displace the bond charge $q = (^2/\epsilon_h)e$ by a distance Δr along the bond direction, where ϵ_h is the homopolar part of the static dielectric constant.[27] This produces a dc dipole moment such that

$$\chi_{\xi\xi}^{dc}\ \Delta E_\xi^{dc} = \mathcal{N} q\ \Delta r. \tag{27}$$

On the other hand, it leads to a change in the charge-transfer energy C_ξ.

$$\Delta C_\xi \cong -f\ e^2\ (Z_A + Z_B)\ \Delta r/r^2 \tag{28}$$

We then have, from Eqs. (24-28),

$$\chi_{\xi\xi\xi}^{(2e)} = {\partial\chi_{\xi\xi}}\Big/{\partial E_\xi^{dc}} = ({\partial\chi_{\xi\xi}}\big/{\partial C_\xi})\ ({\partial C_\xi}\big/{\partial E_\xi^{dc}})$$

$$= (^1/F_{ii}^2)\ [8\, f\ e^2\ (Z_A + Z_B)\ C_\xi/\hbar^2\ q\ d^2 \mathcal{N} \omega_p^2]\ \chi_{ii}^2\ \chi_{ii}^{dc} \tag{29}$$

where $d = r_A + r_B \approx 2r$. To find $\chi^{(2e)}_{ijk}$, we should sum over contributions from all the bonds.

$$\chi^{(2e)}_{ijk} = \sum_{\xi} g_{ijk}\ (\hat{\xi})\ \chi^{(2e)}_{\xi\xi\xi} \tag{30}$$

which reduces to

$$\chi^{(2e)}_{ijk} = G_{ijk}\ (\hat{\xi})\ \chi^{(2e)}_{\xi\xi\xi} \tag{30'}$$

when all bonds are alike. This geometric factor $G_{ijk}\ (\hat{\xi})$ is zero for those $\chi^{(2e)}_{ijk}$ components which must vanish as required by symmetry. When all the quantities in the above expression for $\chi^{(2e)}_{ijk}$ are known for a given crystal, we can then calculate $\chi^{(2e)}_{ijk}$ numerically. Using this model, Levine[24] has calculated $\chi^{(2e)}_{ijk}$ for many crystals of zincblende and wurtzite structures. The agreement with experimental data is excellent, considering the many approximations involved in the calculation.[28]

From Eqs. (29) and (30'), we find

$$\chi^{(2e)}_{ijk} \Big/ \chi^{2}_{ii}\chi^{dc}_{ii} = \Delta_{ijk}$$

$$\Delta_{ijk} = (G_{ijk}\big/F^2_{ii})\ [8\ f\ \ e^2\ (Z_A + Z_B)\big/\hbar^2\ q\ d^2\,\eta\,\omega^2_p]C_{\xi}. \tag{31}$$

This relation was originally suggested by Miller[29] who pointed out that Δ is of the same order of magnitude for a large variety of materials. For all III - V and II - VI compounds, the factor Δ_{ijk}/C_{ξ} should be nearly a constant. The agreement of this assertion with experimental data is again excellent.[24] For large $\chi^{(2e)}_{ijk}$, we must therefore have large linear susceptibility (or small $\bar{E}_g$ as seen from Eq. (24)) and large charge-transfer energy C_{ξ} compatible with small $\bar{E}_g$. In addition, the crystal structure should be in such a way that contributions from various bonds would add rather than subtract.

In the above discussion, we have neglected the local-field correction, which is clearly non-negligible since the valence electrons are fairly localized as seen from the bond picture. However, if we use the observed macroscopic χ_{ii} and χ_{ii}^{dc} in the expression for $\chi_{ijk}^{(2e)}$, then the local-field effect is automatically taken into account.

III. QUADRATIC ELECTRO-OPTICAL EFFECT

We can also derive the microscopic expression for $\underset{\approx}{\chi}^{(3ee)}$ by carrying out the next higher-order perturbation calculation,[20] but again it will be useless for actual evaluation of $\underset{\approx}{\chi}^{(3ee)}$. For the latter purpose, we must use the same approximations discussed in the last section.

Using Levine's approach, we find from Eqs. (24-28)

$$\chi_{\xi\xi\xi\xi}^{(3ee)} = (1/2)\partial^2\chi_{\xi\xi} \Big/ \partial(E_\xi^{dc})^2$$

$$\cong (1/F_{ii})(\chi_{ii}^2 \Big/ \hbar^2\omega_p^2)\,[1 - 4C_\xi^2\,\chi_{ii} \Big/ \hbar^2\omega_p^2]\,(\Delta C_\xi/\Delta E_\xi^{dc})^2$$

$$= (1/F_{ii})^3\,[4\,f\,e^2\,(Z_A + Z_B)/\mathcal{N}q\,d^2\,\hbar^2\,\omega_p^2][1-4C_\xi^2\chi_{ii}/\hbar^2\omega_p^2]\chi_{ii}^2(\chi_{ii}^{dc})^2 \tag{32}$$

where we have neglected the $\partial^2 C_\xi \Big/ \partial(E_\xi^{dc})^2$ term, which is usually small in most crystals.[24] We then obtain

$$\chi_{ijk\ell}^{(3ee)} = G_{ijk\ell}(\hat{\xi})\,\chi_{\xi\xi\xi\xi}^{(3ee)} \tag{33}$$

where $G_{ijk\ell}$ is, again, a geometric factor coming from coordinate transformation. Numerical values of $\chi_{ijk\ell}^{(3ee)}$ calculated from the above expression for several simple crystals also agree well with experimental data.[24] We can also define

$$\Delta_{ijk\ell} = \chi^{(3ee)}_{ijk\ell} \Big/ \chi^2_{ii}(\chi^{dc}_{ii})^2 \tag{33}$$

but now, for the group of semiconductors, $\Delta_{ijk\ell}$ is roughly proportional to $[1 - 4C^2_\xi\, \chi_{ii}/\hbar^2\omega^2_p]$.

For materials in which molecules can rotate and move around, there is also contribution to the quadratic electro-optical effect from molecular reorientation and redistribution by the dc electric field. (or in the quantum picture, population redistribution induced by the field in the ground states). This, in fact, constitutes the major part of the observed quadratic electro-optical effect or Kerr effect in liquid media. Because of the limited scope of this review, we shall not go into detailed theoretical discussion on this subject here, but refer the readers directly to the existing literature.[30]

IV. MAGNETO-OPTICAL EFFECTS

We now consider the effect of a dc magnetic field on the linear dielectric constant ϵ_{ij} or the linear susceptibility χ_{ij}. From Eq. (17), we recognize that the field dependence of χ_{ij} comes in through the wave functions of $|n\rangle$ and $|n'\rangle$, the transition frequencies $\omega_{n'n}$, and the population factors ρ^o_n. In a diamagnetic medium, the states are either fully occupied or empty, and ρ^o_n are unaffected by the dc magnetic field. This change in χ_{ij} induced by field dependence in wave functions and energies is known as the diamagnetic effect.[31] In a paramagnetic medium, however, the populations of the ground states do change as a result of Zeeman perturbation. This population redistribution, which is responsible for paramagnetization, also leads to a change in χ_{ij}, known as the paramagnetic effect.[32] The paramagnetic effect, when present, usually dominates over the diamagnetic counterpart at low temperatures.

Let us first discuss the linear magneto-optical effect. We choose the crystal axes as the coordinate axes. Then, from symmetry argument (Sec. I), $\chi^{(2b)}_{ijk}$ with $i \neq j$ are imaginary and are the only nonvanishing components. The microscopic expressions for χ_{ij} and $\chi^{(2b)}_{ijk}$ with $i \neq j$ can be written as[33,34]

$$\begin{aligned} \chi_{ij} &= (1/2)\,(\chi_{ij} - \chi^{*}_{ij}) \\ &= (Ne^2\omega/\hbar)\sum_g \rho^o_g \sum_n \left[\frac{|(r_+)_{gn}|^2}{(\omega^+_{ng})^2 - \omega^2} - \frac{|(r_-)_{gn}|^2}{(\omega^-_{ng})^2 - \omega^2}\right] \\ &= (Ne^2/\hbar\omega)\sum_g \rho^o_g \sum_n \left[\frac{(\omega^+_{ng})^2|(r_+)_{gn}|^2}{(\omega^+_{ng})^2 - \omega^2} - \frac{(\omega^-_{ng})^2|(r_-)_{gn}|^2}{(\omega^-_{ng})^2 - \omega^2}\right] \end{aligned} \tag{34}$$

$$\begin{aligned} \chi^{(2b)}_{ijk} &= \partial\chi_{ij}\big/\partial H^{dc}_k = (\chi^{(2b)}_{ijk})_D + (\chi^{(2b)}_{ijk})_P \\ (\chi^{(ab)}_{ijk})_D &= (Ne^2\omega/\hbar)\sum_g \rho^o_g\,(\partial/\partial H^{dc}_k)\sum_n\left[\frac{|(r_+)_{gn}|^2}{(\omega^+_{ng})^2 - \omega^2} - \frac{|(r_-)_{gn}|^2}{(\omega^-_{ng})^2 - \omega^2}\right] \\ (\chi^{(2b)}_{ijk})_P &= (Ne^2\omega/\hbar)\sum_g(\partial\rho^o_g/\partial H^{dc}_k)\sum_n\left[\frac{|(r_+)_{gn}|^2}{(\omega^+_{ng})^2 - \omega^2} - \frac{|(r_-)_{gn}|^2}{(\omega^-_{ng})^2 - \omega^2}\right] \end{aligned} \tag{35}$$

where $r_\pm = (r_i \pm r_j)/\sqrt{2}$

$\omega^\pm_{ng} = \omega_{ng} \pm g\mu_B H^{dc}_k$

The above expression for $\chi^{(2b)}_{ijk}$ is again too complicated for numerical evaluation. We are tempted to use the same approximation as used in the discussion of electro-optical effects. However, if $\omega^\pm_{ng}$ are replaced by $\bar{\omega}_{ng}$ in Eqs. (34) and (35), both χ_{ij} and $\chi^{(2b)}_{ijk}$ become zero.[35] This is the result of cancellation between $\sum_n |(r_+)_{gn}|^2$ and $\sum_n |(r_-)_{gn}|^2$. This cancellation is imperfect when the dispersion factors are included in the summation.

For the diamagnetic part, we can write

$$(\chi_{ikj}^{(2b)})_D \cong (Ne^2\omega/\hbar)\sum_g \rho_g^o \sum_n \left\{ \frac{-2\omega_{ng}}{(\omega_{ng}^2-\omega^2)^2}\left[\frac{\partial\omega_{ng}^+}{\partial H_k^{dc}}\,|(r_+)_{gn}|^2 - \frac{\partial\omega_{ng}^-}{\partial H_k^{dc}}\,|(r_-)_{gn}|^2\right]\right.$$
$$\left. + \sum_{n'} \frac{1}{\omega_{ng}^2-\omega^2}\left[(r_+)_{gn}(-M_k)_{nn'}(r_-)_{n'g}-(r_-)_{gn}(-M_k)_{nn'}(r_+)_{n'g}\right]\frac{1}{\hbar\omega_{n'n}}\right\}. \tag{36}$$

The magnitudes of the magnetic-dipole matrix elements $(M_k)_{nn'}$ depend very much on the material system. However, one might expect that the terms involving $(M_k)_{nn'}$ in Eq. (36) are smaller than or, at most, of the same order of magnitude as the other terms. We can then take the approximation

$$(\chi_{ikj}^{(2b)})_D \approx (Ne^2\omega/\hbar)\,\frac{(-2\bar{\omega}_{ng})}{(\bar{\omega}_{ng}^2-\omega^2)^2}\,\overline{\frac{\partial}{\partial H_k^{dc}}(\omega_{ng}^+ - \omega_{ng}^-)}\sum_{g,n}\rho_g^o\,|(r_+)_{gn}|^2$$
$$\approx \chi_{ii}\left[-2\omega\bar{g}\mu_B\Big/(\bar{\omega}_{ng}^2 - \omega^2)\right] \tag{37}$$

where $\bar{g}$ is the mean spectroscopic splitting factor.

For an estimate of the paramagnetic part, let us define the electronic states more explicitly by

$$< n\,| \equiv <\gamma_n,\ J_n|$$

where J_n represents the set of quantum numbers describing the angular part of the state, and γ_n represents the other specifications of the state. We then find from $\sum_n |(r_+)_{gn}|^2 = \sum_n |(r_-)_{gn}|^2$ that

$$\sum_{J_n} |<J_g|r_+|J_n>|^2 = \sum_{J_n} |<J_g|r_-|J_n>|^2 .$$

We can write, with $\Delta_{ng} = \omega_{ng} - \langle\omega_{ng}\rangle_\gamma$, —

$$\sum_{J_n}\left[\frac{|(r_+)_{gn}|^2}{(\omega_{ng}^{+})^2-\omega^2}-\frac{|(r_-)_{gn}|^2}{(\omega_{ng}^{-})^2-\omega^2}\right]\cong\frac{2\langle\omega_{ng}\rangle_\gamma}{(\langle\omega_{ng}\rangle_\gamma^2-\omega^2)^2}\sum_{J_n}\left\{\Delta_{ng}[|(r_+)_{gn}|^2-|(r_-)_{gn}|^2]\right\}$$

$$\approx\left[\langle\omega_{ng}\rangle_\gamma\langle\Delta_{ng}\rangle_\gamma/(\langle\omega_{ng}\rangle_\gamma^2-\omega^2)^2\right]\sum_{J_n}|(r_i)_{gn}|^2. \qquad (38)$$

The expression for $(\chi_{ijk}^{(2b)})_P$ then becomes

$$(\chi_{ijk}^{(2b)})_P\approx(Ne^2\omega/\hbar)\sum_{g,\gamma_n}(\partial\rho_g^o/\partial H_k^{dc})[\langle\omega_{ng}\rangle_\gamma\langle\Delta_{ng}\rangle_\gamma/(\langle\omega_{ng}\rangle_\gamma^2-\omega^2)^2]\sum_{J_n}|(r_i)_{gn}|^2$$

$$\approx\left[\bar{\omega}_{ng}\bar{\Delta}/(\bar{\omega}_{ng}^2-\omega^2)^2\right](Ne^2\omega/\hbar)\sum_{g,n}(\partial\rho_g^o/\partial H_k^{dc})\,|(r_i)_{gn}|^2$$

$$\approx\left[\omega\bar{\Delta}/(\bar{\omega}_{ng}^2-\omega^2)\right]\sum_g(\partial\rho_g^o/\partial H_k^{dc})\,\chi_{ii} \qquad (39)$$

where $\bar{\Delta}$ represents the mean $\langle\Delta_{ng}\rangle_\gamma$. We can imagine that transitions from the ground states to the group of excited states with the same γ_n give rise to a broad absorption band. Then, $\bar{\Delta}$ can be taken approximately as the mean halfwidth of the strong absorption bands. Clearly, the approximation here is ppor, and we should be satisfied with an order-of-magnitude estimate only.

We have $\chi_{ii} = 0.34$ and $\hbar\bar{\omega}_{ng} \approx 7$ ev ~ $\hbar\bar{\omega}_{n'n}$.[27] Then, at $\hbar\omega = 1.1$ eV, we find, from Eq. (36) with $\bar{g} = 2$, $(\chi^{(2b)}_{ijk})_D \approx 2 \times 10^{-10}$/Oe. The Verdet constant for the Faraday rotation is given by

$$V = (2\pi\omega/c)\, \chi^{(2b)}_{ijk}. \qquad (40)$$

We then have $V \approx 7 \times 10^{-5}$ rad/cm - Oe for CdS. The observed Verdet constant for CdS at $\hbar\omega = 1.1$ ev is 3×10^{-5} rad/cm - Oe.[36] As another example, we consider the case of CaF_2 doped with paramagnetic $E\mu^{2+}$ ions. At low temperatures, the paramagnetic term $(\chi^{(2b)}_{ijk})_P$ dominates. Since the absorption edges in both $E\mu^{2+}$ and CdS appear near 4000 Å, we assume the same values of χ_{ii} and $\hbar\bar{\omega}_{ng}$ for $E\mu^{2+}$ as for CdS. The ground states of $E\mu^{2+}$ are the $^8S_{7/2}$ multiplet. We therefore have $\sum_g (\partial\rho^o_g/\partial H) \approx 2\mu_B/kT$. The half-width of a strong band is usually around a few tenths of eV. We set $\bar{\Delta} \approx 0.2$ eV, corresponding to a band width of 3000 cm^{-1}. At $T = 4.2°K$ and $\hbar\omega = 2.26$ eV, we find, from Eq. (38), $(\chi^{(2b)}_{ijk})_P \approx 10^{-7}$/Oe. Then, the Verdet constant for CaF_2:1% $E\mu^{2+}$ is $V \approx 7 \times 10^{-4}$ rad/cm-Oe. The observed value is $V \approx 5 \times 10^{-4}$ rad/cm-Oe.[37] Therefore, in order to have large paramagnetic Verdet constant, we must not only find a crystal with large Zeeman perturbation on the ground-state populations, but should also have the frequency ω close to a strong absorption band.

In general, one can show that under limited conditions, the paramagnetic part of the susceptibility χ_{ij} is proportional to the magnetization $\langle M \rangle$.[32,33]

$$(\chi_{ij})_P = A \sum_m m\rho^o_m = \overrightarrow{A} \langle M \rangle . \qquad (41)$$

The magnetization, and hence $(\chi_{ij})_P$, saturates in the presence of strong field. This is usually the case in ferromagnets. The paramagnetic rotation is always very strong in ferromagnets, but is stronger if ω is close to a strong absorption band in materials.

We can also find the microscopic expression for $\chi^{(3bb)}_{ijk\ell}$ from $\partial^2\chi_{ij}/2\partial H^{dc}_k\partial H^{dc}_\ell$. It is easy to see that for ω sufficiently far away from absorption bands the diamagnetic part of $\chi^{(3bb)}_{ijk\ell}$ is roughly $g\mu_B/\hbar\bar{\omega}_{ng}$ times smaller than $(\chi^{(2b)}_{ijk})_D$, and the paramagnetic part of $\chi^{(3bb)}_{ijk\ell}$ is $g\mu_B/kT\ C_g$ times smaller than $(\chi^{(2b)}_{ijk})_P$. In general, the paramagnetic Voigt effect is given by $[\chi_{ii}(\underset{\sim}{H}^2_{dc}) - \chi_{ii}(0)]_P$, which becomes comparable in order of magnitude with $\chi_{ij}(\underset{\sim}{H}^{dc})$ when the magnetization becomes saturated.

In liquid media, molecules can also be realigned and redistributed by the applied dc magnetic field. Molecular reorientation and redistribution are partially responsible for the observed quadratic magneto-optical effect in liquid. The linear birefringence induced by a magnetic field in liquid is known as the Cotton-Mouton effect. Theoretical treatment on the Cotton-Mouton effect should follow closely to that on the Kerr effect in liquid.

V. CONCLUSION

While symmetry argument is useful to identify the nonvanishing independent components of the $\underset{\approx}{\chi}$ tensors for electro- and magneto-optical effects, a microscopic theory is needed to estimate the magnitudes of these nonvanishing components. Calculations using a simple two-level or two-band model appear to be a good approximation.

Good agreement between theory and experiment for electro-optical coefficients has been particularly encouraging. The same calculations also predict the nonlinear optical coefficients successfully.[19,20,22-24] The approximations used to calculate the Verdet constant for magneto-optical rotation are rather crude, but they do yield order-of-magnitude agreement with the experimental results. The quantitative calculations discussed here break down in the limit where optical absorption becomes appreciable.

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FIGURE CAPTION

Fig. 1. Four equivalent tetrahedral bonds connecting the A (III) atom to the four adjacent B(V) atoms.

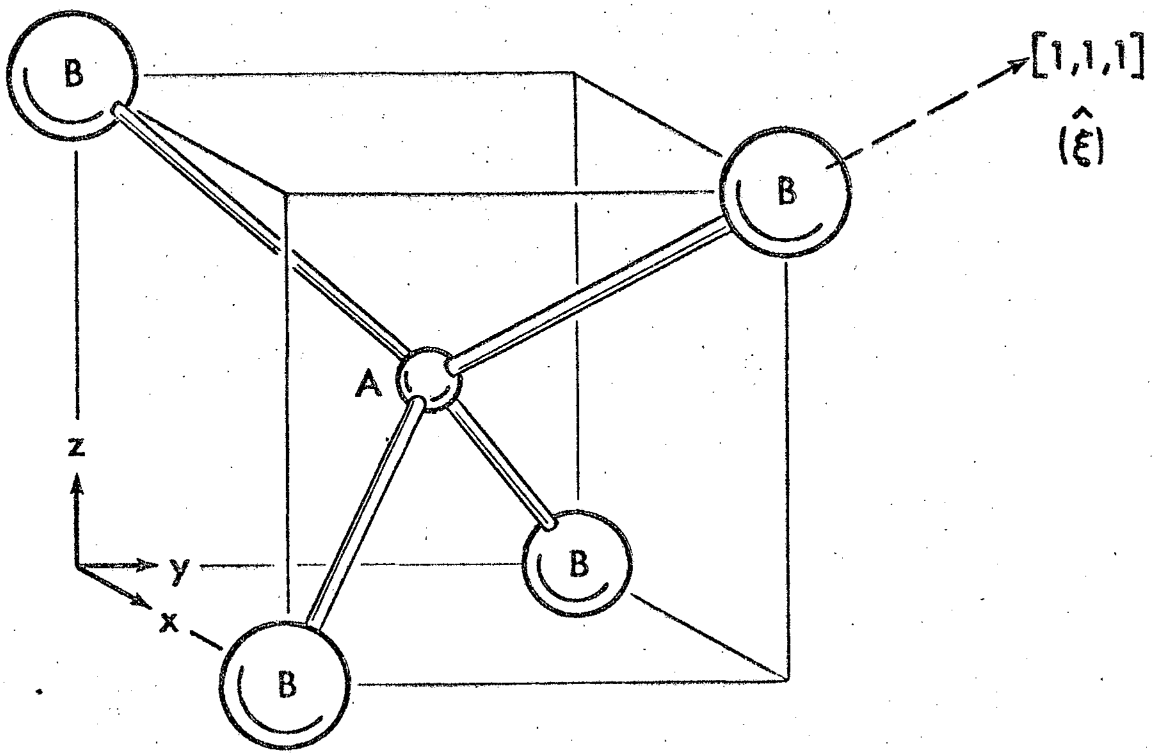

Fig. 1

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