1.6. Classical linear crystal optics

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1.6.1. Introduction

The field of classical crystal optics is an old one, and in the last century, in particular, it was the main subject of interest in the study of crystallography. Since the advent of X-ray diffraction, however, crystal optics tended to fall out of widespread use, except perhaps in mineralogy, where it has persisted as an important technique for the classification and identification of mineral specimens. In more recent times, however, with the growth in optical communications technologies, there has been a revival of interest in the optical properties of crystals, both linear and nonlinear. There are many good books dealing with classical crystal optics, which the reader is urged to consult (Hartshorne & Stuart, 1970; Wahlstrom, 1959; Bloss, 1961). In addition, large collections of optical data on crystals exist (Groth, 1906–1919; Winchell, 1931, 1939, 1951, 1954, 1965; Kerr, 1959). In this chapter, both linear and nonlinear optical effects will be introduced briefly in a generalized way. Then the classical derivation of the refractive index surface for a crystal will be derived. This leads on to a discussion on the practical means by which conventional crystal optics can be used in the study of crystalline materials, particularly in connection with mineralogical study, although the techniques described apply equally well to other types of crystals. Finally, some detailed explanations of certain linear optical tensors will be given.

1.6.2. Generalized optical, electro-optic and magneto-optic effects

When light of a particular cyclic frequency ω is incident on a crystal of the appropriate symmetry, in general an electrical polarization $\mathbf{P}$ may be generated within the crystal. This can be expressed in terms of a power series with respect to the electric vector of the light wave (Nussbaum & Phillips, 1976; Butcher & Cotter, 1990; Kaminow, 1974):

$$P = \sum \varepsilon_o \chi^{(i)} E^i = \varepsilon_o \big(\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \big), \quad (1.6.2.1)$$

where the $\chi^{(i)}$ are susceptibilities of order i. Those working in the field of electro-optics tend to use this notation as a matter of course. The susceptibility $\chi^{(1)}$ is a linear term, whereas the higher-order susceptibilities describe nonlinear behaviour.

However, it is convenient to generalize this concept to take into account other fields (*e.g.* electrical, magnetic and stress fields) that can be imposed on the crystal, not necessarily due to the incident light. The resulting polarization can be considered to arise from many different so-called electro-optic, magneto-optic and photoelastic (elasto-optic) effects, expressed as a series expansion of P_i in terms of susceptibilities $\chi_{ijk\ell\ldots}$ and the applied fields $\mathbf{E}$, $\mathbf{B}$ and T. This can be written in the following way:

$$\begin{aligned} P_i = P_i^0 &+ \varepsilon_o \chi_{ij} E_j^{\omega} + \varepsilon_o \chi_{ij\ell} \nabla_\ell E_j^{\omega} + \varepsilon_o \chi_{ijk} E_j^{\omega_1} E_k^{\omega_2} \\ &+ \varepsilon_o \chi_{ijk\ell} E_j^{\omega_1} E_k^{\omega_2} E_\ell^{\omega_3} + \varepsilon_o \chi_{ijk} E_j^{\omega_1} B_k^{\omega_2} \\ &+ \varepsilon_o \chi_{ijk\ell} E_j^{\omega_1} B_k^{\omega_2} B_\ell^{\omega_3} + \varepsilon_o \chi_{ijk\ell} E_j^{\omega_1} T_{k\ell}^{\omega_2} + \ldots . \end{aligned} \quad (1.6.2.2)$$

Here, the superscripts refer to the frequencies of the relevant field terms and the susceptibilities are expressed as tensor components. Each term in this expansion gives rise to a specific effect that may or may not be observed, depending on the crystal symmetry and the size of the susceptibility coefficients. Note a possible confusion: in the notation $\chi^{(i)}$, i is equal to one less than its rank. It is important to understand that these terms describe various properties, both linear and nonlinear. Those terms that describe the effect purely of optical frequencies propagating through the crystal give rise to *linear* and *nonlinear* optics. In the former case, the input and output frequencies are the same, whereas in the latter case, the output frequency results from sums or differences of the input frequencies. Furthermore, it is apparent that nonlinear optics depends on the intensity of the input field, and so is an effect that is induced by the strong optical field.

If the input electrical fields are static (the term 'static' is used here to mean zero or low frequency compared with that of light), the resulting effects are either linear or nonlinear electrical effects, in which case they are of no interest here. There is, however, an important class of effects in which both static and optical fields are involved: linear and nonlinear electro-optic effects. Here, the use of the terms linear and nonlinear is open to confusion, depending on whether it is the electrical part or the optical part to which reference is made (see for example below in the discussion of the linear electro-optic effect). Similar considerations apply to applied magnetic fields to give linear and nonlinear magneto-optic effects and to applied stresses, the *photoelastic* effects. Table 1.6.2.1 lists the most important effects according to the terms in this series. The susceptibilities are written in the form $\chi(\omega_1; \omega_2, \omega_3, \ldots)$ to indicate the frequency ω_1 of the output electric field, followed after the semicolon by the input frequencies $\omega_1, \omega_2, \ldots$.

Table 1.6.2.1. *Summary of linear and nonlinear optical properties*

Type of polarization term	Susceptibility	Effect
P_i^0	$\chi(0; 0)$	Spontaneous polarization
$\varepsilon_o \chi_{ij} E_j^{\omega}$	$\chi(\omega; \omega)$	Dielectric polarization, refractive index, linear birefringence
$\varepsilon_o \chi_{ij\ell} \nabla_\ell E_j^{\omega}$	$\chi(\omega; \omega)$	Optical rotation (gyration)
$\varepsilon_o \chi_{ijk} E_j^{\omega_1} E_k^{\omega_2}$	$\chi(0; 0, 0)$	Quadratic electric effect
	$\chi(\omega; \omega, 0)$	Linear electro-optic effect or Pockels effect
	$\chi(\omega_1 \pm \omega_2; \omega_1, \omega_2)$	Sum/difference frequency generation, two-wave mixing
	$\chi(\omega; \omega/2, \omega/2)$	Second harmonic generation (SHG)
	$\chi(0; \omega/2, \omega/2)$	Optical rectification
	$\chi(\omega_3; \omega_1, \omega_2)$	Parametric amplification
$\varepsilon_o \chi_{ijk\ell} E_j^{\omega_1} E_k^{\omega_2} E_\ell^{\omega_3}$	$\chi(\omega; 0, 0)$	Quadratic electro-optic effect or Kerr effect
	$\chi(\omega; \omega/2, \omega/2, 0)$	Electric-field induced second harmonic generation (EFISH)
	$\chi(-\omega_1; \omega_2, \omega_3, -\omega_4)$	Four-wave mixing
$\varepsilon_o \chi_{ijk} E_j^{\omega_1} B_k^{\omega_2}$	$\chi(\omega; \omega, 0)$	Faraday rotation
$\varepsilon_o \chi_{ijk\ell} E_j^{\omega_1} B_k^{\omega_2} B_\ell^{\omega_3}$	$\chi(\omega; \omega, 0, 0)$	Quadratic magneto-optic effect or Cotton–Mouton effect
$\varepsilon_o \chi_{ijk\ell} E_j^{\omega_1} T_{k\ell}^{\omega_2}$	$\chi(\omega; \omega, 0)$	Linear elasto-optic effect or photoelastic effect
	$\chi(\omega_1 \pm \omega_2; \omega_1, \omega_2)$	Linear acousto-optic effect

† The sudden death of Keith Cox is deeply regretted. He died in a sailing accident on 27 August 1998 in Scotland at the age of 65.