Chapter 2
Polarization Theory of Nonlinear Medium

This chapter starting from the Maxwell's equations deduces different forms of nonlinear wave equations for the light propagation in the isotropic and anisotropic nonlinear mediums, and in the time and frequency domains; gives the frequency-domain expressions of the polarization and susceptibility of the nonlinear medium; defines the degenerate factor of polarization; introduces the symmetries of nonlinear susceptibility; discusses the relationship between the real part and the imaginary part of susceptibility (K–K relation); points out that the physical meanings of the real part and the imaginary part of third-order nonlinear susceptibility are the nonlinear refractive index and nonlinear absorption coefficient respectively; finally introduces the two kinds of unit systems in nonlinear optics.

2.1 Wave Equations of Nonlinear Medium

2.1.1 Maxwell's Equations for Nonlinear Medium

Under the action of the laser, a medium appears the nonlinear optical effect, which is called the nonlinear medium. When a light wave, as an electromagnetic wave, propagates in the nonlinear medium, it obeys the law depended on the Maxwell equations, in general which can be written as

$$\nabla \times \boldsymbol{E} = -\frac{\partial \boldsymbol{B}}{\partial t}, \tag{2.1.1}$$

$$\nabla \times \boldsymbol{H} = \frac{\partial \boldsymbol{D}}{\partial t} + J, \tag{2.1.2}$$

$$\nabla \cdot \boldsymbol{D} = \rho, \tag{2.1.3}$$

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$$\nabla \cdot \boldsymbol{B} = 0. \tag{2.1.4}$$

It also has the following matter equations:

$$\boldsymbol{D} = \varepsilon_0 \boldsymbol{E} + \boldsymbol{P}, \tag{2.1.5}$$

$$\boldsymbol{B} = \mu_0(\boldsymbol{H} + \boldsymbol{M}), \tag{2.1.6}$$

$$\boldsymbol{J} = \sigma \boldsymbol{E}, \tag{2.1.7}$$

where $\boldsymbol{E}$ and $\boldsymbol{H}$ are denoted the electric field strength and the magnetic field strength, respectively; $\boldsymbol{D}$ and $\boldsymbol{B}$ are denoted the electric induction strength and the magnetic induction strength, respectively; $\boldsymbol{P}$ and $\boldsymbol{M}$ are denoted the electric polarization and magnetic polarization, respectively. For non-ferromagnetic material, the magnetization phenomenon is very week, we can let $\boldsymbol{M} = 0$; ε_0 and μ_0 are denoted the vacuum electric coefficient and the vacuum permeability, respectively; $\boldsymbol{\sigma}$ is the conductivity, strictly speaking, it is a second-order tensor in the anisotropic medium, here approximately is a scalar; $\boldsymbol{J}$ is the conduction current density; $\boldsymbol{\rho}$ is the free charge density, both can be connected each other through the law of conservation of charge:

$$\nabla \cdot \boldsymbol{J} + \frac{\partial \rho}{\partial \boldsymbol{t}} = 0. \tag{2.1.8}$$

For the metal and semiconductor, the conduction current density $\boldsymbol{J}$ and the free charge density $\boldsymbol{\rho}$ these two quantities cannot be neglected, but for the insulator medium, we can assume they are inexistence, then do not consider Eq. (2.1.8). Because the conductivity $\boldsymbol{\sigma}$ is related to the absorption, assuming the linear absorption coefficient is $\boldsymbol{\alpha}$, and then we have relationship $\alpha = \mu_0 \sigma c / n$, so Eq. (2.1.7) should be reserved.

If a strong light (laser) acts on the nonlinear medium, the relationship between $\boldsymbol{P}$ and $\boldsymbol{E}$ is nonlinear, the medium induced $\boldsymbol{P}$ can be spread out into a power series of $\boldsymbol{E}$:

$$\boldsymbol{P} = \varepsilon_0 \boldsymbol{\chi}^{(1)} \cdot \boldsymbol{E} + \varepsilon_0 \boldsymbol{\chi}^{(2)} : \boldsymbol{EE} + \varepsilon_0 \boldsymbol{\chi}^{(3)} \vdots \boldsymbol{EEE} + \cdots, \tag{2.1.9}$$

where $\boldsymbol{\chi}^{(\mathrm{n})}$ is n-order electric susceptibility ($n = 1, 2, 3, \ldots$), which is a $n + 1$-order tensor.

The polarization $\boldsymbol{P}$ can be divided into the linear and nonlinear two parts. The nonlinear part is just the sum of high-order terms of polarization, which is called as nonlinear polarization noted by $\boldsymbol{P}_{\mathbf{NL}}$, that is

$$\boldsymbol{P}_{NL} = \boldsymbol{\varepsilon}_0 \boldsymbol{\chi}^{(2)} : \boldsymbol{EE} + \varepsilon_0 \boldsymbol{\chi}^{(3)} \vdots \boldsymbol{EEE} + \ldots = \boldsymbol{P}^{(2)} + \boldsymbol{P}^{(3)} + \cdots . \tag{2.1.10}$$

Then Eq. (2.1.9) can be expressed as

$$\boldsymbol{P} = \varepsilon_0 \boldsymbol{\chi}^{(1)} \cdot \boldsymbol{E} + \boldsymbol{P}_{NL}. \tag{2.1.11}$$

Substituting Eq. (2.1.11) into Eq. (2.1.5), we obtain

$$\boldsymbol{D} = \varepsilon_0 \boldsymbol{E} + \varepsilon_0 \chi^{(1)} \cdot \boldsymbol{E} + \boldsymbol{P}_{NL} = \boldsymbol{\varepsilon} \cdot \boldsymbol{E} + \boldsymbol{P}_{NL}, \tag{2.1.12}$$

where

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}_0 (1 + \boldsymbol{\chi}^{(1)}) \tag{2.1.13}$$

is the linear dielectric coefficient; in which $\boldsymbol{\chi}^{(1)}$ is linear susceptibility. In the anisotropic medium, $\boldsymbol{\chi}^{(1)}$ and $\boldsymbol{\varepsilon}$ are complex-number second-order tensors. Here we only consider the electric dipole moment approximate, and neglected the action of the electric quadrupole moment and the magnetism dipole moment.

Therefore, Maxwell equations in anisotropic, nonlinear, nonmagnetic medium can be simplified [1] as

$$\nabla \times \boldsymbol{E} = -\mu_0 \frac{\partial \boldsymbol{H}}{\partial t}, \tag{2.1.14}$$

$$\nabla \times \boldsymbol{H} = \frac{\partial \boldsymbol{D}}{\partial t} + \sigma \boldsymbol{E}, \tag{2.1.15}$$

$$\boldsymbol{D} = \boldsymbol{\varepsilon} \cdot \boldsymbol{E} + \boldsymbol{P}_{NL}. \tag{2.1.16}$$

2.1.2 Time-Domain Wave Equation in Anisotropic Nonlinear Medium

In both sides of Eq. (2.1.14) carrying on $\nabla \times$ operation, then substituting Eq. (2.1.15) into it, and using Eq. (2.1.16), finally we obtain

$$\nabla \times \nabla \times \boldsymbol{E} + \mu_0 \sigma \frac{\partial \boldsymbol{E}}{\partial t} + \mu_0 \frac{\partial^2 \boldsymbol{\varepsilon} \cdot \boldsymbol{E}}{\partial t^2} = -\mu_0 \frac{\partial^2 \boldsymbol{P}_{NL}}{\partial t^2}. \tag{2.1.17}$$

This is wave equation for describing the transportation of the light wave in the anisotropic nonlinear medium. In comparison with the linear wave equation, this equation is only added an item on the right side, equivalent to exist a secondary wave source related with polarization $\boldsymbol{P}_{NL}$. The second item on the left is associated with the absorption loss of the medium.

Suppose the medium is lossless, i.e., $\sigma = 0$, and using the formula $c = 1/\sqrt{\mu_0 \varepsilon_0}$, then Eq. (2.1.17) can be written to

$$[\nabla \times (\nabla \times) + \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\boldsymbol{\varepsilon}\cdot]\,\boldsymbol{E}(\boldsymbol{r},t) = -\frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\boldsymbol{P}_{NL}(\boldsymbol{r},t). \tag{2.1.18}$$

This is time-domain wave equation in the non-absorption anisotropic nonlinear medium.

The light field strength $\boldsymbol{E}$ and nonlinear polarization $\boldsymbol{P}_{NL}$ in Eqs. (2.1.17) and (2.1.18) are the function of the time and position. In order to solve the equation and find the optical field strength $\boldsymbol{E}$, we mast firstly find out the nonlinear polarization $\boldsymbol{P}_{NL}$.

2.1.3 Time-Domain Wave Equation in Isotropic Nonlinear Medium

Assuming that the nonlinear medium is an non-absorption, homogeneous, isotopic medium, in Eq. (2.1.18) $\nabla \cdot \boldsymbol{E} = 0$, then $\nabla \times \nabla \times \boldsymbol{E} = \nabla(\nabla \cdot \boldsymbol{E}) - \nabla^2 \boldsymbol{E} = -\nabla^2 \boldsymbol{E}$; the original $\boldsymbol{\varepsilon}$ is a tensor, if the light wave with the amplitude of $\boldsymbol{E}$ is a plane wave and a transverse wave, its component paralleled to propagation direction can be neglected, so the tensor $\boldsymbol{\varepsilon}$ can be written to scalar quantity $\boldsymbol{\varepsilon}$. Using formula $n = \sqrt{\varepsilon/\varepsilon_0}$, thus Eq. (2.1.18) becomes

$$\nabla^2 \boldsymbol{E}(\boldsymbol{r},t) - \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\boldsymbol{E}(\boldsymbol{r},t) = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\boldsymbol{P}_{NL}(\boldsymbol{r},t). \tag{2.1.19}$$

This is a time-domain wave equation for the plane light wave propagates in a non-absorption and isotopic nonlinear medium, which is an inhomogeneous second-order differential equation, it is difficulty to solve, in general, it needs approximately simplification treatment, the slow amplitude approximation is an used way. By using this method, the second-order differential equation will become a first-order differential equation.

In order to simplify, assuming a monochromic plane wave field propagates along z-direction, and the light field strength and the nonlinear polarization are denoted as a product of amplitude factor and phase factor, respectively:

$$\boldsymbol{E}(\mathrm{r},t) = \boldsymbol{E}(z,t)e^{i(kz-\omega t)}, \tag{2.1.20}$$

$$\boldsymbol{P}_{NL}(\boldsymbol{r},t) = \boldsymbol{P}_{NL}(z,t)e^{i(k'z-\omega t)}. \tag{2.1.21}$$

Substituting Eqs. (2.1.20) and (2.1.21) into Eq. (2.1.19), in which the various items have following differential coefficients of $\boldsymbol{E}(z,t)$ and $\boldsymbol{P}_{NL}(z,t)$, respectively:

$$\nabla^2 \boldsymbol{E}(z,t) = \left[\left(\frac{\partial^2}{\partial z^2} + i2k\frac{\partial}{\partial z} - k^2\right)|\boldsymbol{E}(z,t)|\right] e^{i(kz-\omega t)}, \tag{2.1.22}$$

$$\frac{\partial^2}{\partial t^2}\boldsymbol{E}(z,t) = \left[\left(\frac{\partial^2}{\partial t^2} - i2\omega\frac{\partial}{\partial t} - \omega^2\right)|\boldsymbol{E}(z,t)|\right] e^{i(kz-\omega t)}, \tag{2.1.23}$$

$$\frac{\partial^2}{\partial t^2}\boldsymbol{P}_{NL}(z,t) \cong -\omega^2 |\boldsymbol{P}_{NL}(z,t)| e^{i(k'z-\omega t)}. \tag{2.1.24}$$

Suppose the variation of field strength is very slow in the space distant within the scope of light wavelength and within the time scope of optical frequency; i.e., satisfy the following space and time conditions of the slowly varying field amplitude approximation [2]

$$\left|\frac{\partial^2 \boldsymbol{E}(z,t)}{\partial z^2}\right| << \left|k\frac{\partial \boldsymbol{E}(z,t)}{\partial z}\right| \text{ and } \left|\frac{\partial^2 \boldsymbol{E}(z,t)}{\partial t^2}\right| << \left|\omega\frac{\partial \boldsymbol{E}(z,t)}{\partial t}\right|. \tag{2.1.25}$$

Substituting Eqs. (2.1.22)–(2.1.24) into Eq. (2.1.19), using the slowly varying amplitude approximation condition of Eq. (2.1.25), omitting the items with second-order differential coefficients for the space and time, and using $k = (\omega/c)n$ and $v = c/n$, therefore, we obtain the following Eq. [3]:

$$\frac{\partial \boldsymbol{E}(z,t)}{\partial z} + \frac{1}{v}\frac{\partial \boldsymbol{E}(z,t)}{\partial t} = \frac{i\omega}{2\varepsilon_0 cn}\boldsymbol{P}_{NL}(z,t)e^{i\Delta kz}, \tag{2.1.26}$$

where $\Delta k = k' - k$, k and k' are the wave vectors of original light field and polarization field, respectively. Equation (2.1.26) is the time-domain wave equation when the monochromic plane light wave propagates in non-absorption and isotopic nonlinear medium, and the optical field strength $\boldsymbol{E}(z,t)$ satisfies the space and time slowly varying amplitude approximation condition. If the light wave is a continuous wave, or a light pulse with a wide pulse width, in Eq. (2.1.26) $v = c/n$ is the phase velocity of light wave; if the light wave is a short pulse (for example is a picosecond pulse), it is not a monochromic wave, we can regard as a wave packet, the form of time-domain nonlinear wave equation is same as Eq. (2.1.26), in which optical field amplitude $\boldsymbol{E}(z,t)$ should express as an integral of time. In this case, the group velocity of wave pocket should denoted by $v = d\omega/dk$.

2.1.4 Frequency-Domain Wave Equation in Anisotropic Nonlinear Medium

The anisotropic-medium time-domain nonlinear wave Eq. (2.1.18) can be changed to frequency-domain formation. For this purpose we should pass through Fourier transform, to spread $\boldsymbol{E}(\boldsymbol{r}, t)$ and $\boldsymbol{P}_{NL}(\boldsymbol{r}, t)$ into the sum of multiple monochromic plane waves, and write each monochromic wave to be the product of amplitude and phase two factors, then we have

$$\boldsymbol{E}(\boldsymbol{r}, t) = \sum_i \boldsymbol{E}_i(\mathbf{k}_i, \omega_i) = \sum_i \boldsymbol{E}_i e^{\mathrm{i}(\mathbf{k}_i \boldsymbol{r} - \omega_i t)}, \tag{2.1.27}$$

$$\begin{aligned}\boldsymbol{P}_{NL}(\boldsymbol{r}, t) &= \sum_{n\geq 2} \boldsymbol{P}^{(n)}(\boldsymbol{r}, t) = \sum_{n\geq 2}\sum_i \boldsymbol{P}_i^{(n)}(\boldsymbol{k}_i', \omega_i) \\ &= \sum_i \boldsymbol{P}_i^{NL}(\boldsymbol{k}_i', \omega_i) = \sum_i \boldsymbol{P}_i^{NL}(\boldsymbol{k}_i', \omega_i) e^{\mathrm{i}(\boldsymbol{k}_i' \boldsymbol{r} - \omega_i t)},\end{aligned} \tag{2.1.28}$$

where ω is the angular frequency of light wave, $\boldsymbol{k}$ and $\boldsymbol{k}'$ are wave vectors of the light field and the polarization field of the monochromic plane wave, respectively. To substitute Eqs. (2.1.27) and (2.1.28) into Eq. (2.1.18), and omit the summation mark and ordinal number i, we can obtain

$$[\nabla \times (\nabla\times) - \frac{\omega^2}{\varepsilon_0 c^2}\boldsymbol{\varepsilon}\cdot]\boldsymbol{E}(\boldsymbol{k}, \omega) = \frac{\omega^2}{\varepsilon_0 c^2}\boldsymbol{P}_{NL}(\boldsymbol{k}', \omega). \tag{2.1.29}$$

This is the frequency-domain wave equation for the monochromic plane wave propagating in a non-absorption anisotropic nonlinear medium.

2.1.5 Frequency-Domain Wave Equation in Isotopic Nonlinear Medium

Assuming that the medium is isotopic and homogeneous; $\boldsymbol{E}(\boldsymbol{k}, \omega)$ is denoted the light field strength of the monochromic plane wave, which is a transverse wave, i.e., the component paralleled to $\boldsymbol{K}$ can be neglected, so that in wave Eq. (2.1.29) $\nabla \cdot \boldsymbol{E} = 0$, and $\nabla \times \nabla \times \boldsymbol{E} = -\nabla^2 \boldsymbol{E}$, the tensor $\boldsymbol{\varepsilon}$ can be written to the scalar $\boldsymbol{\varepsilon}$, further using the relations $k = k_0 n$, $k_0 = \omega/c$ and $n = \sqrt{\varepsilon/\varepsilon_0}$, then we obtain

$$\nabla^2 \boldsymbol{E}(\boldsymbol{k}, \omega) + k^2 \boldsymbol{E}(\boldsymbol{k}, \omega) = -\frac{k_0^2}{\varepsilon_0}\boldsymbol{P}_{NL}(\boldsymbol{k}', \omega). \tag{2.1.30}$$

This is frequency-domain wave equation for the monochromic plane wave propagating in a non-absorption isotopic nonlinear medium. This is an inhomogeneous second-order differential equation, it is difficult to directly solve. We can take a simplified deal with by using the slowly varying amplitude approximate method as follows.

Considering that the monochrome plane wave propagates along z-direction, its amplitude varies with z, but no change with time. The light field strength and nonlinear polarization are respectively denoted by

$$\boldsymbol{E}(\boldsymbol{k}, \omega) = \boldsymbol{E}(z, \omega)e^{\mathrm{i}(kz-\omega t)}, \tag{2.1.31}$$

$$\boldsymbol{P}_{NL}(\boldsymbol{k}, \omega) = \boldsymbol{P}_{NL}(z, \omega)e^{i(k\prime z-\omega t)}. \tag{2.1.32}$$

where $\boldsymbol{k}$ and $\boldsymbol{k}^{'}$ are wave vectors of the original light field and polarization field respectively. $\boldsymbol{E}(z, \omega)$ and $\boldsymbol{P}_{NL}(z, \omega)$ denote the field amplitudes and nonlinear polarization respectively. Substituting Eqs. (2.1.31) and (2.1.32) into Equation (2.1.30), the first item of left of Eq. (2.1.30) is

$$\nabla^2\boldsymbol{E}(z, \omega) = \left(\frac{\partial^2}{\partial z^2} + i2k\frac{\partial}{\partial z} - k^2\right)\boldsymbol{E}(z, \omega)e^{i(kz-\omega t)}. \tag{2.1.33}$$

So the items containing coefficient k^2 in Eq. (2.1.30) can be eliminated, Eq. (2.1.30) becomes

$$\left(\frac{\partial^2}{\partial z^2} + i2k\frac{\partial}{\partial z}\right)\boldsymbol{E}(z, \omega)e^{i(kz-\omega t)} = -\frac{k_0^2}{\varepsilon_0}\boldsymbol{P}_{NL}(z, \omega)(z)e^{i(k'z-\omega t)}. \tag{2.1.34}$$

Suppose that the light field strength satisfies the space slowly varying field amplitude approximation condition:

$$\left|\frac{\partial^2\boldsymbol{E}(z, \omega)}{\partial z^2}\right| < < \left|k\frac{\partial \boldsymbol{E}(z, \omega)}{\partial z}\right|, \tag{2.1.35}$$

the items having second derivative of light field strength in Eq. (2.1.34) can be omitted, and to combine the exponent factors in both side of equation, Eq. (2.1.34) can be written to

$$\frac{\partial \boldsymbol{E}(z, \omega)}{\partial z} = \frac{ik_0^2}{2\varepsilon_0 k}\boldsymbol{P}_{NL}(z, \omega)e^{\mathrm{i}\Delta kz}, \tag{2.1.36}$$

where

$$\Delta k = k' - k, \tag{2.1.37}$$

where $\boldsymbol{k}$ and k' are wave vectors of the original light field and the polarization field, respectively.

Because $k = (\omega/c)n$ and $k_0 = \omega/c$, then Eq. (2.1.36) also can be expressed as

$$\frac{\partial \boldsymbol{E}(z,\omega)}{\partial z} = \frac{i\omega}{2\varepsilon_0 cn}\boldsymbol{P}_{NL}(z,\omega)e^{i\Delta kz}. \tag{2.1.38}$$

Equation (2.1.38) is a frequency-domain wave equation for a monochromic plane light wave propagating along z-direction in the isotopic, uniform and lossless nonlinear medium under the condition of slowly varying amplitude approximation. It is a first-order differential equation, relatively easy to be solved. If know the nonlinear polarization $\boldsymbol{P}_{NL}(z,\omega)$, we can obtain the solution of light field strength $\boldsymbol{E}(z,\omega)$. In this book, all of the investigative nonlinear optical processes will describe and explain by using this simple first-order differential equation.

Firstly, using the nonlinear coupling wave equations we can solve the multi-wave mixing nonlinear optical problem. Generally speaking, for n-order nonlinear polarization effect, we can list $n+1$ nonlinear coupling wave Equations similar to Eq. (2.1.38), simultaneous solving these $n+1$ equations, we can find the $n+1$ light field strengths with different frequency, thus obtain the law of energy mutual transformation among these light fields.

For example, for the second-order nonlinear optical effect, there are 2 original light fields at two different frequencies and 1 new generated polarization field, it requires 3 coupling wave equations, we can simultaneous solve these 3 coupling wave equations to obtain the 3 field strengths; For the third-order nonlinear optical effect, there are 3 original light fields and 1 new generated polarization field, it requires 4 coupling equations, then we can simultaneous solve out the 4 field strengths.

If existing absorption in the medium, according to Eq. (2.1.17), $\sigma \neq 0$, we can obtain the slowly varying amplitude approximation frequency-domain wave equation considering the absorption for the propagation of the monochromatic light wave along z-direction:

$$\frac{\partial \boldsymbol{E}(z,\omega)}{\partial z} + \frac{\alpha}{2}\boldsymbol{E}(z,\omega) = \frac{i\omega}{2\varepsilon_0 cn}\boldsymbol{P}_{NL}(z,\omega)e^{i\Delta kz}, \tag{2.1.39}$$

where $\alpha = \mu_0 \sigma c/n$ is the linear absorption coefficient of medium.

2.2 Polarization and Susceptibility of Nonlinear Medium

2.2.1 Frequency-Domain Expressions of Polarization and Susceptibility

1. **Frequency-Domain Expressions in Anisotropic Medium**

Under the action of light field, the polarization phenomenon is generated in an anisotropic medium. What is relationship between the polarization $\boldsymbol{P}$ and the light electric field strength $\boldsymbol{E}$? At first, we are going to investigate the causal relationship in time between $\boldsymbol{P}$ and $\boldsymbol{E}$ [4], and then discuss the frequency-domain expressions of polarization $\boldsymbol{P}$ and susceptibility $\boldsymbol{\chi}$ in the case of the linear optics and different order nonlinear optics phenomena.

(1) **In Linear Polarization Case**

The induced electric polarization of medium $d\boldsymbol{P}^{(1)}(t)$ at the moment t is generated by the light electric filed strength $\boldsymbol{E}(t_1)$ before the moment $t_1 = t - dt_1$, $d\boldsymbol{P}^{(1)}(t)$ and $\boldsymbol{E}(t_1)$ has the following direct-ratio relation in the time interval dt_1:

$$d\boldsymbol{P}^{(1)}(t) = \varepsilon_0 \boldsymbol{\chi}^{(1)}(t - t_1) \cdot \boldsymbol{E}(t_1) dt_1. \tag{2.2.1}$$

Considering the contribution of $\boldsymbol{E}(t_1)$ to $\boldsymbol{P}^{(\mathbf{1})}(t)$ in all time before the moment t, we have

$$\boldsymbol{P}^{(1)}(t) = \int_{-\infty}^{\infty} \varepsilon_0 \boldsymbol{\chi}^{(1)}(t - t_1) \cdot \boldsymbol{E}(t_1) dt_1. \tag{2.2.2}$$

Actual, when $t_1 < t$, $\boldsymbol{E}(t_1)$ has no contribution to $\boldsymbol{P}^{(1)}(t)$, i.e., $\boldsymbol{\chi}^{(1)}(t - t_1) = 0$.

In order to further get the relationship between $\boldsymbol{P}$ and $\boldsymbol{E}$ in the frequency-domain, we take the Fourier transform of $\boldsymbol{E}(t_1)$ and $\boldsymbol{P}^{(1)}(t)$, i.e.,

$$\boldsymbol{E}(t_1) = \int_{-\infty}^{\infty} \boldsymbol{E}(\omega) e^{-i\omega t_1} d\omega, \tag{2.2.3}$$

$$\boldsymbol{P}^{(1)}(t) = \int_{-\infty}^{\infty} \boldsymbol{P}(\omega)^{(1)} e^{-i\omega t} d\omega. \tag{2.2.4}$$

To substitute Eqs. (2.2.3) and (2.2.4) into Eq. (2.2.2), and eliminate the integral sign, we obtain the frequency-domain expression:

$$\boldsymbol{P}^{(1)}(\omega) = \varepsilon_0 \boldsymbol{\chi}^{(1)}(\omega) \cdot \boldsymbol{E}(\omega), \tag{2.2.5}$$

where

$$\boldsymbol{\chi}^{(1)}(\omega) = \int_{-\infty}^{\infty} \boldsymbol{\chi}^{(1)}(t - t_1) e^{i\omega(\mathrm{t} - \mathrm{t}_1)} dt_1. \tag{2.2.6}$$

where $\boldsymbol{\chi}^{(1)}(\omega)$ is a linear polarization tensor, it is a second-order tensor with 9 tensor elements, that is

$$\boldsymbol{\chi}^{(1)}(\omega) = \begin{bmatrix} \chi_{11}^{(1)}(\omega) & \chi_{12}^{(1)}(\omega) & \chi_{13}^{(1)}(\omega) \\ \chi_{21}^{(1)}(\omega) & \chi_{22}^{(1)}(\omega) & \chi_{23}^{(1)}(\omega) \\ \chi_{31}^{(1)}(\omega) & \chi_{32}^{(1)}(\omega) & \chi_{33}^{(1)}(\omega) \end{bmatrix}. \tag{2.2.7}$$

In the rectangular coordinate system, the every element of linear polarization tensor can be expressed by its index, i.e.,

$$\boldsymbol{\chi}^{(1)}(\omega) = \begin{bmatrix} XX & XY & XZ \\ YX & YY & YZ \\ ZX & ZY & ZZ \end{bmatrix}. \tag{2.2.8}$$

(2) **In Nonlinear Polarization Case**

As mention in the previous section, the polarization $\boldsymbol{P}$ can spread as the power series of $\boldsymbol{E}$, in the frequency domain the $\boldsymbol{P}$ can be expressed as

$$\boldsymbol{P}(\omega) = \boldsymbol{P}^{(1)}(\omega) + \boldsymbol{P}^{(2)}(\omega) + \boldsymbol{P}^{(3)}(\omega) + \cdots . \tag{2.2.9}$$

Similar to Eqs. (2.2.5) and (2.2.6), the second-order nonlinear polarization and the second-order nonlinear susceptibility can be expressed as

$$\boldsymbol{P}^{(2)}(\omega) = \varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega; \omega_1, \omega_2) : \boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2), \tag{2.2.10}$$

$$\boldsymbol{\chi}^{(2)}(\omega) = \int_{-\infty}^{\infty} \boldsymbol{\chi}^{(2)}(t - t_1, t - t_2) e^{i[\omega_1(t-t_1) + \omega_2(t-t_2)]} dt_1 dt_2. \tag{2.2.11}$$

$\boldsymbol{\chi}^{(2)}$ is called second-order polarization tensor, it is a third-order tensor with 27 tensor elements:

$$\boldsymbol{\chi}^{(2)}(\omega) = \begin{bmatrix} XXX & XYY & XZZ & XYZ & XZY & XZX & XXZ & XXY & XYZ \\ YXX & YYY & YZZ & YYZ & YZY & YZX & YXZ & YXY & YYZ \\ ZXX & ZYY & ZZZ & ZYZ & ZZY & ZZX & ZXZ & ZXY & ZYZ \end{bmatrix}. \tag{2.2.12}$$

In a similar way, the third-order nonlinear polarization and the third-order nonlinear susceptibility can be expressed as respectively

$$\boldsymbol{P}^{(3)}(\omega) = \varepsilon_0 \boldsymbol{\chi}^{(3)}(\omega; \omega_1, \omega_2, \omega_3) \vdots \boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2)\boldsymbol{E}(\omega_3), \tag{2.2.13}$$

$$\boldsymbol{\chi}^{(3)}(\omega) = \int_{-\infty}^{\infty} \boldsymbol{\chi}^{(3)}(t-t_1, t-t_2, t-t_3) e^{i[\omega_1(t-t_1)+\omega_2(t-t_2)+\omega_3(t-t_3)]} dt_1 dt_2 dt_3. \tag{2.2.14}$$

$\boldsymbol{\chi}^{(3)}$ is called third-order polarization tensor, it is a four-order tensor with 81 tensor elements.

In the similar way, n-order nonlinear polarization and n-order nonlinear susceptibility can be expressed as

$$\boldsymbol{P}^{(n)}(\omega) = \varepsilon_0 \boldsymbol{\chi}^{(n)}(\omega; \omega_1, \omega_2, \ldots, \omega_n) \vdots \boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2), \ldots, \boldsymbol{E}(\omega_n), \tag{2.2.15}$$

where $\boldsymbol{\chi}^{(n)}$ is a n+1-order tensor, sign of "$\vdots$" is denoted the multiplication of n +1-order tensor.

$$\boldsymbol{\chi}^{(n)}(\omega) = \int_{-\infty}^{\infty} \boldsymbol{\chi}^{(n)}(t-t_1, t-t_2, \ldots, t - t_n) e^{i[\omega_1(t-t_1)+\omega_2(t-t_2)+\ldots+\omega_n(t-t_n)]} dt_1 dt_2 \ldots dt_n. \tag{2.2.16}$$

It is worth noting that in the bracket of above every order susceptibility is inserted a semicolon, according to the regulation of this book, after the semicolon are original light fields at the frequencies of $\omega_1, \omega_2, \ldots, \omega_n$; before the semicolon is the polarization field at the frequency of ω. According to the energy conservation law, the frequency of polarization field is the sum of frequencies of all original fields:

$$\omega = \omega_1 + \omega_2 + \cdots + \omega_n. \tag{2.2.17}$$

2. **Rectangular Coordinate Frequency-Domain Expressions in Anisotropic Medium**

Below we provide the frequency-domain expressions by rectangular coordinate component for each order polarizations in anisotropic medium. The polarization-filed frequency ω is a sum of the original-field frequencies, $\omega = \omega_1 + \omega_2 + \omega_3 + \cdots$.

$$P_\mu^{(1)}(\omega) = \sum_\alpha \varepsilon_0 \chi_{\mu\alpha}^{(1)}(\omega; \omega) E_\alpha(\omega), \tag{2.2.17}$$

$$P_\mu^{(2)}(\omega) = \sum_{\alpha\beta} \varepsilon_0 \chi_{\mu\alpha\beta}^{(2)}(\omega; \omega_1, \omega_2) E_\alpha(\omega_1) E_\beta(\omega_2), \tag{2.2.18}$$

$$P_{\mu}^{(3)}(\omega) = \sum_{\alpha\beta\gamma} \varepsilon_0 \chi_{\mu\alpha\beta\gamma}^{(3)}(\omega; \omega_1, \omega_2, \omega_3) E_{\alpha}(\omega_1) E_{\beta}(\omega_2) E_{\gamma}(\omega_3), \tag{2.2.19}$$

$$P_{\mu}^{(n)}(\omega) = \sum_{\alpha\beta\gamma} \varepsilon_0 \chi_{\mu\alpha\beta\gamma}^{(n)}(\omega; \omega_1, \omega_2, \ldots, \omega_n) E_{\alpha}(\omega_1) E_{\beta}(\omega_2) \ldots E_{\gamma}(\omega_n), \tag{2.2.20}$$

式中 $\mu, \alpha, \beta, \gamma, \ldots = x, y, z.$

3. **Frequency-Domain Expressions in Isotopic Medium**

For uniform isotopic nonlinear medium, the light field strength $\boldsymbol{E}(\omega)$ and the polarization $\boldsymbol{P}(\omega)$ are complex number vectors, and the susceptibility $\boldsymbol{\chi}$ is a complex number scalar. The each-order polarization can be expressed as follows.

Linear polarization:

$$\boldsymbol{P}^{(1)}(\omega) = \varepsilon_0 \boldsymbol{\chi}^{(1)}(\omega; \omega) \boldsymbol{E}(\omega). \tag{2.1.21}$$

Second-order nonlinear polarization:

$$\boldsymbol{P}^{(2)}(\omega) = \varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega; \omega_1, \omega_2) \boldsymbol{E}(\omega_1) \boldsymbol{E}(\omega_2). \tag{2.1.22}$$

Third-order nonlinear polarization:

$$\boldsymbol{P}^{(3)}(\omega) = \varepsilon_0 \boldsymbol{\chi}^{(3)}(\omega; \omega_1, \omega_2, \cdots \omega_3) \boldsymbol{E}(\omega_1) \boldsymbol{E}(\omega_2) \boldsymbol{E}(\omega_3). \tag{2.1.23}$$

The n-order nonlinear polarization:

$$\boldsymbol{P}^{(n)}(\omega) = \varepsilon_0 \boldsymbol{\chi}^{(n)}(\omega; \omega_1, \omega_2, \ldots, \omega_n) \boldsymbol{E}(\omega_1) \boldsymbol{E}(\omega_2) \ldots \boldsymbol{E}(\omega_n). \tag{2.1.24}$$

When you write the the expressions of polarization, you should determine the frequences of each light field participated in the designated nonlinear optical process and write out correct susceptibility expressions. If in the incident light electric fields having conjugate complex number of field, we should add a minus sign in the front of corresponding frequency inside the bracket of susceptibility.

2.2.2 Degeneration Factor of Polarization

Assuming the incident light field is consisted by a series monochromic plane waves at frequencies $\omega_1, \omega_2, \cdots \omega_n$. The electric field strength of each monochromic plane wave at frequency $\omega_i (i = 1, 2, 3 \cdots)$ is a complex number, in general it can be written to a sum of complex number and its conjugate complex number (c.c). Therefore the total incident light electric field strength $\boldsymbol{E}(\boldsymbol{r}, t)$ can be expressed as

$$\boldsymbol{E}(\boldsymbol{r},t) = \sum_{\mathrm{i}} \boldsymbol{E}(\omega_{\mathrm{i}}) e^{-i(\omega_{\mathrm{i}} t - \boldsymbol{k}_i \cdot \boldsymbol{r})} + c.c.. \tag{2.2.25}$$

The n-order polarization at the frequency of $\omega = \omega_1 + \omega_2 + \cdots + \omega_n$ is induced by n original light fields at frequencies of $\omega_1, \omega_2, \cdots, \omega_n$. Actually, the original light fields may include several light fields at the same frequency, i.e., existing the frequency degeneracy. Because the frequency degeneracy and the symmetry of susceptibility, in the rectangular coordinate component expression for the anisotropic nonlinear medium should add a coefficient D, which is called degeneration factor, so the rectangular coordinate component expression of n-order polarization is given by

$$P_{\mu}^{(n)}(\omega) = D \sum_{\alpha\beta\cdots\gamma} \varepsilon_0 \boldsymbol{\chi}_{\mu\alpha\beta\cdots\gamma}^{(n)}(\omega; \omega_1, \omega_2, \omega_3, \cdots, \omega_n) E_{\alpha}(\omega_1) E_{\beta}(\omega_2) \cdots E_{\gamma}(\omega_n), \tag{2.2.26}$$

For the isotopic nonlinear medium, if all the light fields propagate along the z-direction, the n-order polarization expression in the frequency degeneracy case also need add the degeneration factor, namely

$$\boldsymbol{P}^{(n)}(z,\omega) = D\varepsilon_0 \boldsymbol{\chi}^{(n)}(\omega; \omega_1, \omega_2, \ldots, \omega_n) \boldsymbol{E}(z,\omega_1) \boldsymbol{E}(z,\omega_2) \ldots \boldsymbol{E}(z,\omega_n), \tag{2.2.27}$$

It can be proved that, if the light field strength is expressed as Eq. (2.2.25), the formula of degeneration factor is given by [5]

$$D = \frac{n!}{m!}, \tag{2.2.28}$$

where n is the order number of nonlinear polarization, m is the frequency degenerate number of original light fields. In this book the light field strength is all expressed as Eq. (2.2.25), so the Eq. (2.2.28) is used to calculate the degeneration factor in this book.

In some literature, considering the relation $\boldsymbol{E}(t) = \boldsymbol{E}_0 \cos \omega t = \frac{1}{2} \boldsymbol{E}_0 (e^{\mathrm{i}\omega \mathrm{t}} + e^{-\mathrm{i}\omega t})$, the light field strength is expressed as

$$\boldsymbol{E}(\boldsymbol{r},t) = \frac{1}{2} \sum_{\mathrm{i}} \boldsymbol{E}(\omega_{\mathrm{i}}) e^{-i(\omega_{\mathrm{i}} t - \boldsymbol{k}_i \cdot \boldsymbol{r})} + c.c. \tag{2.2.29}$$

It can be proved that in this case the formula of degeneration factor should be written to

$$D = 2^{1-n} \left(\frac{n!}{m!} \right). \tag{2.2.30}$$

Same as above, here n is the order number of nonlinear polarization, m is the frequency degenerate number of original light fields.

Table 2.1 list the susceptibility expressions and corresponding two kinds of degeneration factors for various nonlineal optical effects. In the susceptibility expressions, after the semicolon are the frequencies of the incident (original) light fields, $\omega_1, \omega_2, \cdots, \omega_n$; before the semicolon is the frequency of the generated (polarization) field $\omega = \omega_1 + \omega_2 + \cdots + \omega_n$, the negative frequency delegates that its light field is a conjugate complex of the light field at positive frequency.

Table 2.1 Susceptibility expressions and two kinds of degeneration factors for nonlineal optical effects

Nonlinear optical process	Order (n)	Susceptibility	$D = n!/m!$	$D = 2^{1-n}(n!/m!)$
Linear absorption	1	$\chi^{(1)}(\omega;\omega)$	1	1
Linear refraction	1	$\chi^{(1)}(\omega;\omega)$	1	1
Electrooptical effect	2	$\chi^{(2)}(\omega;\omega,0)$	2	1
Frequency doubling effect	2	$\chi^{(2)}(2\omega;\omega,\omega)$	1	1/2
Sum frequency effect	2	$\chi^{(2)}(\omega_3;\omega_1,\omega_2)$	2	1
Difference frequency effect	2	$\chi^{(2)}(\omega_2;\omega_3,-\omega_1)$	2	1
Triple frequency harmonic	3	$\chi^{(3)}(3\omega;\omega,\omega,\omega)$	1	1/4
Single-photon nonlinear refraction	3	$\chi^{(3)}(\omega;\omega,-\omega,\omega)$	3	3/4
Single-photon nonlinear absorption	3	$\chi^{(3)}(\omega;\omega,-\omega,\omega)$	3	3/4
Two-photon nonlinear absorption	3	$\chi^{(3)}(\omega_1;\omega_2,-\omega_2,\omega_1)$	6	3/2
Self-phase modulation optical Kerr effect	3	$\chi^{(3)}(\omega;\omega,-\omega,\omega)$	3	3/4
Cross-phase modulation optical Kerr effect	3	$\chi^{(3)}(\omega;\omega_{\mathrm{p}},-\omega_p,\omega)$	6	3/2
Four wave mixing	3	$\chi^{(3)}(\omega_4;\omega_1,\omega_2,\omega_3)$	6	3/2
Degenerate four wave mixing	3	$\chi^{(3)}(\omega;\omega,-\omega,\omega)$	3	3/4
Degenerate four wave mixing back phase conjugation	3	$\chi^{(3)}(\omega_c;\omega_1,-\omega_2,\omega_p)$	6	3/2
Degenerate four wave mixing forward phase conjugation	3	$\chi^{(3)}(\omega_c;\omega_1,\omega_2,-\omega_p)$	6	3/2
Stoks stimulated Raman scattering	3	$\chi^{(3)}(\omega_{\mathrm{s}};\omega_{\mathrm{p}},-\omega_p,\omega_s)$	6	3/2
Anti-stoks stimulated Raman scattering	3	$\chi^{(3)}(\omega_{as};\omega_p,\omega_p,-\omega_s)$	3	3/4

2.2.3 Symmetry of Susceptibility Tensor

There are some universal symmetric relationships among tensor elements of nonlinear susceptibility tensor. These symmetric relationships reflect the real-number character of polarization and the symmetry of medium structure. If you want to know the certification of these symmetries in detail, you can refer to relater reference materials [4, 6–8]. Here we just give a brief introduction.

1. **Authenticity Condition**

Linear susceptibility tensor of medium can be expressed as the form of Eq. (2.2.6):

$$\boldsymbol{\chi}^{(1)}(\omega) = \int_{-\infty}^{\infty} \boldsymbol{\chi}^{(1)}(t-t_1)e^{i\omega(t-t_1)}dt_1.$$

Taking its conjugate complex number, we obtain

$$\boldsymbol{\chi}^{(1)*}(\omega) = \int_{-\infty}^{\infty} \boldsymbol{\chi}^{(1)}(t-t_1)e^{-i\omega(\mathrm{t}-\mathrm{t}_1)}dt_1 = \boldsymbol{\chi}^{(1)}(-\omega). \tag{2.2.31}$$

In a similar way, we can prove that the tensor elements of each-order nonlinear susceptibility have following characteristics:

$$\boldsymbol{\chi}^{(2)*}_{ijk}(\omega;\omega_1,\omega_2) = \boldsymbol{\chi}^{(2)}_{jki}(\omega_1;-\omega_2,\omega) = \boldsymbol{\chi}^{(2)}_{kji}(\omega_2;-\omega_1,\omega), \tag{2.2.32}$$

$$\begin{aligned}\boldsymbol{\chi}^{(3)*}_{ijkl}(\omega;\omega_1,\omega_2,\omega_3) &= \boldsymbol{\chi}^{(3)}_{jkli}(\omega_1;-\omega_2,-\omega_3,\omega)\\ &= \boldsymbol{\chi}^{(3)}_{kijl}(\omega_2;\omega,-\omega_1,-\omega_3)\\ &= \boldsymbol{\chi}^{(3)}_{lijk}(\omega_3;\omega,-\omega_1,-\omega_2),\end{aligned} \tag{2.2.33}$$

$$\ldots\ldots$$

Above relationships guarantee the characteristics that the each susceptibility is a real number, so it is called the authenticity condition. If the frequency of original optical field is far from the resonance frequency of medium, the medium is know as non-dispersion and lossless, we can remove out the symbol of conjugate complex number "*" in Eqs. (2.2.32)–(2.2.33).

1. **Symmetry of Intrinsic Substitution**

It can be proved that susceptibility tensor has following intrinsic symmetry of frequency substitution: if do not change the location of polarization-field frequency, when the sequence of any two frequencies of original optical field mutually interchanges, corresponding two tensor elements are same. For example, for second-order and third-order nonlinear effects we have:

$$\chi_{ijk}^{(2)}(\omega;\omega_m,\omega_n) = \chi_{ikj}^{(2)}(\omega;\omega_n,\omega_m), \tag{2.2.34}$$

$$\begin{aligned}\chi_{ijkl}^{(3)}(\omega;\omega_m,\omega_n,\omega_q) &= \chi_{ikjl}^{(3)}(\omega;\omega_n,\omega_m,\omega_q)\\ &= \chi_{ijlk}^{(3)}(\omega;\omega_m,\omega_q,\omega_n)\\ &= \chi_{ilkj}^{(3)}(\omega;\omega_q,\omega_n,\omega_m).\end{aligned} \tag{2.2.35}$$

2. **Symmetry of Complete Substitution**

If the frequency of original optical field far from the resonance frequency of medium, the medium is know as non-dispersion and lossless, there is the symmetry of complete substitution, i.e., the frequency of the polarization field can interchange place with any frequency of original field, for example, the second-order and the third-order nonlinear optical effects have:

$$\chi_{ijk}^{(2)}(\omega;\omega_m,\omega_n) = \chi_{jik}^{(2)}(\omega_m;\omega,\omega_n) = \chi_{kji}^{(2)}(\omega_n;\omega_m,\omega), \tag{2.2.36}$$

$$\begin{aligned}\chi_{ijkl}^{(3)}(\omega;\omega_m,\omega_n,\omega_q) &= \chi_{jikl}^{(3)}(\omega_m;\omega,\omega_n,\omega_q)\\ &= \chi_{kjil}^{(3)}(\omega_n;\omega_m,\omega,\omega_q)\\ &= \chi_{ljki}^{(3)}(\omega_q;\omega_m,\omega_n,\omega).\end{aligned} \tag{2.2.37}$$

In the above tensor elements of susceptibility, interchange the frequency in any order, the numerical value of tensor elements keeps no change; this feature is called complete-substitution symmetry.

3. **Time Reversion Symmetry**

According to the real number character of nonlinear polarization, it is can be proved that any tensor element possesses following characteristic:

$$\chi_{ll_1l_2\cdots l_n}^{(n)}(\omega;\omega_1,\omega_2,\ldots,\omega_n) = \chi_{ll_1l_2\cdots l_n}^{(n)}(-\omega;-\omega_1,-\omega_2,\ldots,-\omega_n). \tag{2.2.38}$$

4. **Spatial Structure Symmetry**

We mentioned previous, the susceptibility is a 3-D space tensor. In which $\boldsymbol{\chi}^{(1)}$ is a second-order tensor, with 9 tensor elements; $\boldsymbol{\chi}^{(2)}$ is a third-order tensor, with 27 tensor elements; $\boldsymbol{\chi}^{(3)}$ is a four-order tensor, with 81 tensor elements. Because the structure of nonlinear medium (such as nonlinear crystal) has symmetry (rotate symmetry and translation symmetry, etc.), it makes the part of tensor elements to be zero, and there exists specific relationships among some tensor elements, to lead total number of independent tensor elements of nonlinear susceptibility tensor depletes.

According to the symmetry of the crystal, the crystal can be divided into 7 systems of crystallization: triclinic crystal system, monoclinic crystal system, orthorhombic crystal system, quadratic crystal system, trigonal crystal system, hexagonal crystal system, cubic crystal system (or isotopic medium). People already found the susceptibility tensor form of these 7 crystal systems and their 32 crystal classes. Visible, the symmetry is higher; the number of the non-zero tensor element and independent tensor element is less.

For example, for second-order tensor $\boldsymbol{\chi}^{(1)}$, its orthorhombic crystal system and cubic crystal system (or isotopic medium) only have 3 and 1 independent tensor element respectively:

$$\boldsymbol{\chi}^{(1)}(\omega)=\begin{bmatrix} XX & 0 & 0 \\ 0 & YY & 0 \\ 0 & 0 & ZZ \end{bmatrix}, \qquad \boldsymbol{\chi}^{(1)}(\omega)=\begin{bmatrix} XX & 0 & 0 \\ 0 & XX & 0 \\ 0 & 0 & XX \end{bmatrix}.$$

Orthorhombic crystal system Cubic crystal system and isotopic medium

For another example, for third-order tensor $\boldsymbol{\chi}^{(2)}$, its crystal class 222 (D_2) of orthorhombic crystal system and crystal class $\bar{4}3\,m$ (T_d) of cubic crystal system, they only have 6 and 1 independent tensor element respectively:

$$\boldsymbol{\chi}^{(2)}(\omega)=\begin{bmatrix} 0 & 0 & 0 & \mathrm{XYZ} & \mathrm{XZY} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & YZX & YXZ & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & ZXY & ZYZ \end{bmatrix}$$

Crystal class 222 (D_2) of orthorhombic crystal system

$$\boldsymbol{\chi}^{(2)}(\omega)=\begin{bmatrix} 0 & 0 & 0 & XYZ & XYZ & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & XYZ & XYZ & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & XYZ & XYZ \end{bmatrix}$$

Crystal class $\bar{4}3\,m$ (T_d) of cubic crystal system

Now we study the characteristics of susceptibility of the medium with centre (inversion) symmetry, So called centre symmetry, it is $\boldsymbol{P}$ and $\boldsymbol{E}$ should change to revers direction under inverse conversion of coordinate $\{x,y,z\}\to\{-x,-y,-z\}$ to keep the formula of polarization invariability. That is to say, for the following general formula of n-order polarization:

$$\boldsymbol{P}^{(n)}(\omega)=\varepsilon_0\boldsymbol{\chi}^{(n)}(\omega;\omega_1,\omega_2,\ldots,\omega_n)\,\vdots\,\boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2)\ldots\boldsymbol{E}(\omega_n), \tag{2.2.39}$$

when making conversion of coordinates $\{x, y, z\} \rightarrow \{-x, -y, -z\}$, in left side of formula the vector $\boldsymbol{P}$ becomes $-\boldsymbol{P}$, and in the right side of formula every vector $\boldsymbol{E}$ becomes $-\boldsymbol{E}$, so the Eq. (2.2.39) becomes

$$\boldsymbol{P}'^{(n)}(\omega) = (-1)^{n+1}[\varepsilon_0 \boldsymbol{\chi}'^{(n)}(\omega;\omega_1,\omega_2,\ldots,\omega_n) \vdots \boldsymbol{E}'(\omega_1)\boldsymbol{E}'(\omega_2)\ldots\boldsymbol{E}'(\omega_n)]. \tag{2.2.40}$$

In order to maintain the form of Eq. (2.2.40) same as that of Eq. (2.2.39), we should require that when $n = 2, 4, \ldots$ (even number), $\boldsymbol{P}'^{(n)} = 0$, i.e., $\boldsymbol{\chi}'^{(n)}(\omega;\omega_1, \omega_2,\ldots,\omega_n) = 0$. Namely even-order susceptibility is zero for the medium with centre symmetry. If we only consider the nonlinear effect up to third-order, for the medium with centre symmetry, it is nonexistence of second-order nonlinear effect, only existence of third-order nonlinear effect.

2.3 Real Part and Imaginary Part of Susceptibility

2.3.1 Relation Between Real Part and Imaginary Part of Susceptibility (K–K Relation)

The susceptibility $\chi(\omega)$ is a function of frequency, in general it is a complex number, can be expressed as

$$\chi(\omega) = \chi'(\omega) + i\chi''(\omega). \tag{2.3.1}$$

Between the real part and the imaginary part of linear susceptibility has following relationships (the derivation of the formula see Appendix 2.A):

$$\chi'(\omega) = \frac{1}{\pi} P.V. \int_{-\infty}^{\infty} \frac{\chi''(\omega')}{\omega' - \omega} d\omega', \tag{2.3.2}$$

$$\chi''(\omega) = -\frac{1}{\pi} P.V. \int_{-\infty}^{\infty} \frac{\chi'(\omega')}{\omega' - \omega} d\omega', \tag{2.3.3}$$

The integral in Eqs. (2.3.2) and (2.3.3) is Cauchy's principle value integral, namely when integral removing the singular point $\omega' = \omega$. Equations (2.3.2) and (2.3.3) are famous *Kramers-Kronig* dispersion relation, in short K–K relation [9, 10]. From K–K relation we can see that as long as know any one of the real part and the imaginary part of the susceptibility as a function of frequency, we can through above relationship to find out another one.

According to the substitution symmetry of susceptibility $\chi(-\omega) = \chi^*(\omega)$ and Eq. (2.3.1), we can rewrite the K–K relation to be:

$$\chi'(\omega) = \frac{2}{\pi} P.V. \int_0^\infty \frac{\omega' \chi''(\omega')}{\omega'^2 - \omega^2} d\omega'', \tag{2.3.4}$$

$$\chi''(\omega) = -\frac{2\omega}{\pi} P.V. \int_0^\infty \frac{\chi'(\omega')}{\omega'^2 - \omega^2} d\omega'. \tag{2.3.5}$$

Because integral just in positive frequency range, the K–K relations as form of Eqs. (2.3.4) and (2.3.5) are more accord with the physical significance.

K–K relation is derived from the linear susceptibility, for the linear optical system it is always correct, but only a part of processes in nonlinear optical system comply with K–K relation. For instance, second harmonic effect, third harmonic effect, four-wave mixing (except degenerate four-wave mixing), cross-phase modulation Kerr effect (except self-phase modulation Kerr effect) etc. (see Ref. [10]).

2.3.2 *Physical Significance of Real Part and Imaginary Part of Susceptibility*

1. **Relation of Linear Susceptibility with Linear Refractive Index and Linear Absorption Coefficient**

Now we investigate that a monochrome plane wave at frequency of ω propagates in an isotopic medium along z-direction to generate the polarization. Suppose the light field denoted by

$$\boldsymbol{E}(z, \omega) = \boldsymbol{E}(z) e^{\mathrm{i}(kz - \omega t)} + c.c., \tag{2.3.6}$$

where $\boldsymbol{k}$ is a complex number wave vector, its real part k' is related with the refractive index of medium; the imaginary part k'' is related with the absorption coefficient of medium, namely

$$k = k' + ik'' = k_0 n_0 + i\frac{\alpha_0}{2}, \tag{2.3.7}$$

where $k_0 = \omega/c$ is the wave vector in vacuum; n_0 and α_0 are linear refractive index and linear absorption of medium respectively (α_0 is absorption coefficient for light power, so it is divided by 2).

Considering the linear polarization effect in far from the resonance situation, using the definition of electric induction strength, we obtain

$$\boldsymbol{D} = \varepsilon_0 \boldsymbol{E} + \boldsymbol{P}^{(1)} = \varepsilon_0 \boldsymbol{E} + \varepsilon_0 \chi^{(1)}(\omega)\boldsymbol{E} = [\varepsilon_0 + \varepsilon_0 \chi^{(1)}(\omega)]\boldsymbol{E} = \varepsilon \boldsymbol{E}, \tag{2.3.8}$$

where ε_0 is the dielectric coefficient in vacuum; $\varepsilon = \varepsilon_0 + \varepsilon_0 \chi^{(1)}(\omega)$ is the complex linear dielectric coefficient of medium; $\chi^{(1)}(\omega)$ is complex linear susceptibility of medium, which can be divided into the real and the imaginary two parts: $\chi^{(1)}(\omega) = \chi^{(1)\prime}(\omega) + i\chi^{(1)\prime\prime}(\omega)$. Using relation $\varepsilon' = \varepsilon_0[1 + \chi^{(1)\prime}(\omega)]$, then $\boldsymbol{\varepsilon}$ can be expressed as

$$\varepsilon = \varepsilon_0 + \varepsilon_0 \chi^{(1)\prime}(\omega) + i\varepsilon_0 \chi^{(1)\prime\prime}(\omega) = \varepsilon' + i\varepsilon_0 \chi^{(1)\prime\prime}(\omega) = \varepsilon'[1 + i\frac{\varepsilon_0}{\varepsilon'}\chi^{(1)\prime\prime}(\omega)]. \tag{2.3.9}$$

To use the linear refractive index $n_0 = \sqrt{\varepsilon'/\varepsilon_0}$, Eq. (2.3.9) can be written as

$$\varepsilon = n_0^2 \varepsilon_0 [1 + i\frac{\chi^{(1)\prime\prime}(\omega)}{n_0^2}]. \tag{2.3.10}$$

Further use complex linear refractive index $n = \sqrt{\varepsilon/\varepsilon_0}$ and light velocity in vacuum $c = 1/\sqrt{\mu_0 \varepsilon_0}$, the complex wave vector of medium can be written as

$$k = \frac{\omega}{c} n = \omega\sqrt{\mu_0 \varepsilon}. \tag{2.3.11}$$

Substituting Eq. (2.3.10) into Eq. (2.3.11), we obtain

$$k = k_0 n_0 \left(1 + i\frac{\chi^{(1)\prime\prime}(\omega)}{n_0^2}\right)^{\frac{1}{2}}, \tag{2.3.12}$$

In the bracket of left of Eq. (2.3.12), the model of second item is much smaller than 1, so the bracket factor can be spread to Taylor's series, after that approximately taking front two items, we obtain

$$k \approx k_0 n_0 \left[1 + i\frac{\chi^{(1)\prime\prime}(\omega)}{2n_0^2}\right] = k_0 n_0 + i\frac{k_0}{2n_0}\chi^{(1)\prime\prime}(\omega). \tag{2.3.13}$$

To compare the Eq. (2.3.13) with Eq. (2.3.7), and use of $n_0 = \sqrt{\varepsilon'/\varepsilon_0}$ and $\varepsilon' = \varepsilon_0[1 + \chi^{(1)\prime}(\omega)]$, we obtain

$$n_0 = [1 + \chi^{(1)\prime}(\omega)]^{\frac{1}{2}} \approx 1 + \frac{1}{2}\chi^{(1)\prime}(\omega), \tag{2.3.14}$$

$$\alpha_0 = \frac{k_0}{n_0}\chi^{(1)''}(\omega) = \frac{\omega}{cn_0}\chi^{(1)''}(\omega). \quad (2.3.15)$$

We can see that, the linear reflective index and the linear absorption coefficient of the medium are linearly related with the real part and the imaginary part of first-order susceptibility, respectively.

2. **Relation of Third-Order Nonlinear Susceptibility with Nonlinear Refractive Index and Nonlinear Absorption Coefficient**

We suppose the medium is a third-order nonlinear medium; the input laser is monochromatic plane wave as expressed by Eq. (2.3.6), the light field $\boldsymbol{E}(z)$ can be solved by using the slowly varying amplitude approximation nonlinear wave Eq. (2.1.38). Here let $\Delta k = k' - k = 0$. Equation (2.1.38) becomes

$$\frac{\partial \boldsymbol{E}(z)}{\partial z} = \frac{i\omega}{2\varepsilon_0 cn_0}\boldsymbol{P}_{\mathrm{NL}}(z). \quad (2.3.16)$$

The third-order nonlinear polarization (for example, Kerr effect) can be expressed as

$$\boldsymbol{P}_{NL}(z) = \boldsymbol{P}^{(3)}(z) = 3\varepsilon_0\chi^{(3)}(\omega)|\boldsymbol{E}(z)|^2\boldsymbol{E}(z). \quad (2.3.17)$$

Using $\chi^{(3)} = \chi^{(3)'}(\omega) + i\chi^{(3)''}(\omega)$, Eq. (2.3.17) becomes

$$\boldsymbol{P}_{\mathrm{NL}}(z) = 3\varepsilon_0[\chi^{(3)'}(\omega)|\boldsymbol{E}(z)|^2 + i\chi^{(3)''}(\omega)|\boldsymbol{E}(z)|^2]\boldsymbol{E}(z). \quad (2.3.18)$$

Substituting Eq. (2.3.18) into Eq. (2.3.16), we obtain

$$\frac{\partial \boldsymbol{E}(z)}{\partial z} = \frac{i3\omega}{2cn_0}[\chi^{(3)'}(\omega)|\boldsymbol{E}(z)|^2 + i\chi^{(3)''}(\omega)|\boldsymbol{E}(z)|^2]\boldsymbol{E}(z). \quad (2.3.19)$$

Using $I = \frac{1}{2}\varepsilon_0 cn_0|\boldsymbol{E}(z)|^2$, Eq. (2.3.19) becomes

$$\frac{\partial \boldsymbol{E}(z)}{\partial z} = i3\left[k_0\frac{\chi^{(3)'}(\omega)}{\varepsilon_0 cn_0^2}I + i\frac{\omega\chi^{(3)''}(\omega)}{\varepsilon_0 c^2 n_0^2}I\right]\boldsymbol{E}(z). \quad (2.3.20)$$

Setting

$$k_{NL} = 3k_0\frac{\chi^{(3)'}(\omega)}{\varepsilon_0 cn_0^2}I + i3\frac{\omega\chi^{(3)''}(\omega)}{\varepsilon_0 c^2 n_0^2}I, \quad (2.3.21)$$

Equation (2.3.20) becomes

$$\frac{\partial \boldsymbol{E}(z)}{\partial z} = ik_{NL}\boldsymbol{E}(z). \tag{2.3.22}$$

The solution of Eq. (2.3.22) is

$$\boldsymbol{E}(z) = \boldsymbol{E}(0)e^{ik_{NL}z}. \tag{2.3.23}$$

From the definition Eq. (2.3.6) of light field

$$\boldsymbol{E}(z,\omega) = \boldsymbol{E}(z)e^{i(kz-\omega t)} = \boldsymbol{E}(0)e^{ik_{NL}z}e^{i(kz-\omega t)} = \boldsymbol{E}(0)e^{ik_T z}e^{-i\omega t} \tag{2.3.24}$$

where $k_T = k + k_{NL}$ is the total wave vector, it is a complex number, can be divided into the real part and the imaginary part, the real part is corresponding to the total refractive index n; the imaginary part is corresponding to the total absorption α:

$$k_T = k'_T + ik''_T = n + i\frac{\alpha}{2}. \tag{2.3.25}$$

The each of total refractive index and total absorption coefficient can be divided into linear and nonlinear two parts, namely

$$n = n_0 + \Delta n, \tag{2.3.26}$$

$$\alpha = \alpha_0 + \Delta\alpha. \tag{2.3.27}$$

Substituting Eqs. (2.3.26) and (2.3.27) into Eq. (2.3.25), we obtain

$$k_T = k_0 n_0 + k_0\Delta n + i\frac{\alpha_0}{2} + i\frac{\Delta\alpha}{2}. \tag{2.3.28}$$

Using Eq. (2.3.21), the total wave vector can be written to the sum of linear and nonlinear two parts:

$$k_T = k + k_{NL} = k_0 n_0 + i\frac{\alpha_0}{2} + 3k_0\frac{\chi^{(3)\prime}(\omega)}{\varepsilon_0 c n_0^2}I + i3\frac{\omega\chi^{(3)\prime\prime}(\omega)}{\varepsilon_0 c^2 n_0^2}I. \tag{2.3.29}$$

To compare Eqs. (2.3.28) and (2.3.29), then we obtain the expressions of nonlinear refractive index Δn and the nonlinear absorption coefficient $\Delta\alpha$:

$$\Delta n = \frac{3}{\varepsilon_0 c n_0^2}\chi^{(3)\prime}(\omega)I, \tag{2.3.30}$$

$$\Delta\alpha = \frac{6\omega}{\varepsilon_0 c^2 n_0^2}\chi^{(3)\prime\prime}(\omega)I. \tag{2.3.31}$$

From Eqs. (2.3.30) to (2.3.31) we can see that for the third-order nonlinear medium, its nonlinear refractive index depends on the real part of third-order susceptibility and is proportional to the light intensity; its nonlinear absorption coefficient depends on the imaginary part of third-order susceptibility and is also proportional to the light intensity.

2.3.3 Relation Between Nonlinear Refractive Index and Nonlinear Absorption Coefficient

If K–K relation is applicative to a certain third-order nonlinear process, from Eqs. (2.3.30) and (2.3.31) we can obtain the relation between the real part of susceptibility $\chi^{(3)\prime}(\omega)$ and the nonlinear refractive index $\Delta n(\omega)$, and the relation between the imaginary part of susceptibility $\chi^{(3)\prime\prime}(\omega)$ and the nonlinear absorption coefficient $\Delta\alpha(\omega)$, respectively. Substituting them into Eq. (2.3.4) of K–K relation, thus we get the relation between the nonlinear refractive index $\Delta n(\omega)$ and the nonlinear absorption coefficient $\Delta\alpha(\omega)$:

$$\Delta n(\omega) = \frac{c}{\pi} P.V. \int_0^{\infty} \frac{\Delta\alpha(\omega')}{\omega'^2 - \omega^2} d\omega'. \tag{2.3.32}$$

Because the nonlinear refractive index of medium is very difficult to measure directly, often pass through measuring nonlinear absorption coefficient to indirectly determine the nonlinear refractive index. If measured the linear absorption spectrum of a nonlinear medium and its nonlinear absorption spectrum under a high power light, from the difference of these two spectrums to obtain $\Delta\alpha(\omega')$, then we can using Eq. (2.3.32) to calculate the nonlinear refractive index at the frequency of ω, $\Delta n(\omega)$. Then we can also use Eq. (2.3.30) to reversely calculate the real part of the third-order susceptibility at that frequency $\chi^{(3)\prime}(\omega)$.

Review Questions of Chapter 2

1. From Maxwell equations to deduce the time-domain wave equation for the light wave propagates in the anisotropic nonlinear medium and the isotopic nonlinear medium.
2. To deduce frequency-domain wave equation for the monochromic plane light wave in the anisotropic nonlinear medium and the isotopic nonlinear medium.
3. To deduce the steady-state wave equation and dynamic equation for monochromic plane light wave in isotopic medium under the slowly-varying-amplitude approximation.
4. Write down the general frequency-domain expression of nonlinear polarization. What is degeneration factor? List several examples of second and third-order nonlinear effects, and write their nonlinear polarization expressions (including degeneration factors).

5. What kinds of symmetries do the nonlinear polarization tensor has? Why the mediums with centre symmetry have no the second-order nonlinearity, only have the third-order nonlinearity?
6. Write down the expression of relation between real part and imaginary part of linear susceptibility (K–K relation). What high order nonlinear processes can apply K–K relation?
7. For linear medium, what are the relationships of the refractive index and absorption coefficient with the real part and the imaginary part of the susceptibility respectively? For third-order nonlinear medium, what is the relationship between the nonlinear refractive index and the nonlinear absorption coefficient?
8. In nonlinear optics, there are two systems of units: International system (MKS//SI) and Gaussian system (mgs/esu). How to distinguish the basic formula in these two systems and how to convert the units between two systems?

Appendix A: Derivation of K–K Relation [9, 10]

K–K relation is obtained at first from linear system. Firstly we consider the mathematic property of linear susceptibility. The light frequency can be regard as a complex number quantity, in the complex number plane of frequency ω' we integral to linear susceptibility $\chi^{(1)}(\omega')$ as follows.

$$\int_{-\infty}^{\infty} \frac{\chi^{(1)}(\omega')d\omega}{\omega' - \omega}. \tag{2.A.1}$$

Considering there is a singular point at point of $\omega' = \omega$. In order to avoid that singular point, we integral along a loop on the upper half of ω' complex number plane ($\omega' \geq 0$), as shown in Fig. 2.1; then take the limitation of $R \to \infty$, $\varepsilon \to 0$, the integral of Eq. (2.A.1) can be finished. According to Cauchy's theorem, because it has no singular point on the closed loop, the integral should be zero. We can explain physically like this: for the real frequency ω, the susceptibility $\chi(\omega)$ is measurable, so that it is limited, the integral to it is convergent.

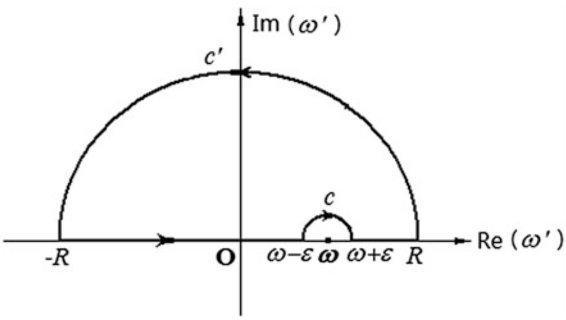

Fig. 2.1 Field of integration used for derivation of K–K relation

The integral of loop can be divided into following four segments: the semicircle c′ around the original point from −***R*** to ***R***; the semicircle c around point ω from $\omega-\varepsilon$ to $\omega+\varepsilon$; and the two straight lines along the real axis of ω': from –***R*** to $\omega-\varepsilon$ and from $\omega+\varepsilon$ to ***R***, i.e.,

$$\int_{c'} \frac{\chi^{(1)}(\omega')}{\omega'-\omega} d\omega' + \int_{c} \frac{\chi^{(1)}(\omega')}{\omega'-\omega} d\omega' + \left[\int_{-R}^{\omega-\varepsilon} \frac{\chi^{(1)}(\omega')d\omega'}{\omega'-\omega} + \int_{\omega+\varepsilon}^{R} \frac{\chi^{(1)}(\omega')d\omega'}{\omega'-\omega} \right] = 0. \tag{2.A.2}$$

For the first item, the integral of c′ tends to zero with $|R| \to \infty$, that is because when $|R| \to \infty$, $|\omega'|$ increases, $\chi(\omega')/|\omega'|$ tens to zero. For the second item, in the integral of ***c***, assuming $\omega' = \omega + \varepsilon e^{\mathrm{i}\varphi}$, when $\varepsilon \to 0$, the integral becomes

$$\lim_{\varepsilon\to 0} \int_{c} \frac{\chi^{(1)}(\omega')}{\omega'-\omega} d\omega' = \lim_{\varepsilon\to 0} \int_{\pi}^{0} \frac{\chi^{(1)}(\omega+\varepsilon e^{\mathrm{i}\varphi}) i e^{\mathrm{i}\varphi}}{\varepsilon e^{\mathrm{i}\varphi}} d\varphi = -i\pi\chi^{(1)}(\omega). \tag{2.A.3}$$

For the third item, when $R \to \infty$, it is Cauchy-principal-value integral:

$$\left[\int_{-\infty}^{\omega-\varepsilon} \frac{\chi^{(1)}(\omega')d\omega'}{\omega'-\omega} + \int_{\omega+\varepsilon}^{\infty} \frac{\chi^{(1)}(\omega')d\omega'}{\omega'-\omega} \right] = P.V. \int_{-\infty}^{\infty} \frac{\chi^{(1)}(\omega')d\omega'}{\omega'-\omega}. \tag{2.A.4}$$

In the condition of $R \to \infty$, $\varepsilon \to 0$, substituting Eqs. (2.A.3) and (2.A.4) into Eq. (2.A.2), we obtain

$$\chi^{(1)}(\omega) = -\frac{i}{\pi} P.V. \int_{-\infty}^{\infty} \frac{\chi^{(1)}(\omega')}{\omega'-\omega} d\omega'. \tag{2.A.5}$$

To substrate $\chi^{(1)}(\omega) = \chi^{(1)\prime}(\omega) + i\chi^{(1)\prime\prime}(\omega)$ and $\chi^{(1)}(\omega') = \chi^{(1)\prime}(\omega') + i\chi^{(1)\prime\prime}(\omega')$ into Eq. (2.A.5) respectively, and the real part and imaginary part respectively equal, then we obtain K–K relation expressions as same as Eqs. (2.3.2) and (2.3.3) in the case of linear polarization.

$$\chi^{(1)\prime}(\omega) = \frac{1}{\pi} P.V. \int_{-\infty}^{\infty} \frac{\chi^{(1)\prime\prime}(\omega')}{\omega'-\omega} d\omega', \tag{2.A.6}$$

$$\chi^{(1)\prime\prime}(\omega) = -\frac{1}{\pi} P.V. \int_{-\infty}^{\infty} \frac{\chi^{(1)\prime}(\omega')}{\omega'-\omega} d\omega'. \tag{2.A.7}$$

According to $\chi^{(1)}(-\omega') = \chi^{(1)}(\omega')^*$, $\chi^{(1)\prime}(\omega')$ is the even function of ω', i.e., $\chi^{(1)\prime}(-\omega') = \chi^{(1)\prime}(\omega')$; and $\chi^{(1)\prime\prime}(\omega')$ is odd function of ω', i.e., $\chi^{(1)\prime\prime}(-\omega') = -\chi^{(1)\prime\prime}(\omega')$, therefore Eqs. (2.A.6) and (2.A.7) can be written as

$$\begin{aligned}\chi^{(1)\prime}(\omega) &= \frac{1}{\pi}P.V.\left[\int_{-\infty}^{0} \frac{\chi^{(1)\prime\prime}(\omega')}{\omega'-\omega}d\omega' + \int_{0}^{\infty} \frac{\chi^{(1)\prime\prime}(\omega')}{\omega'-\omega}d\omega'\right] \\ &= \frac{1}{\pi}P.V.\left[\int_{0}^{\infty} \frac{\chi^{(1)\prime\prime}(\omega')}{\omega'+\omega}d\omega' + \int_{0}^{\infty} \frac{\chi^{(1)\prime\prime}(\omega')}{\omega'-\omega}d\omega'\right] \\ &= \frac{2}{\pi}P.V.\int_{0}^{\infty} \frac{\omega'\chi^{(1)\prime\prime}(\omega')}{(\omega'^2-\omega^2)}d\omega',\end{aligned} \tag{2.A.8}$$

$$\begin{aligned}\chi^{(1)\prime\prime}(\omega) &= -\frac{1}{\pi}P.V.\left[\int_{-\infty}^{0} \frac{\chi^{(1)\prime}(\omega')}{\omega'-\omega}d\omega' + \int_{0}^{\infty} \frac{\chi^{(1)\prime}(\omega')}{\omega'-\omega}d\omega'\right] \\ &= \frac{1}{\pi}P.V.\left[\int_{0}^{\infty} \frac{\chi^{(1)\prime}(\omega')}{\omega'+\omega}d\omega' - \int_{0}^{\infty} \frac{\chi^{(1)\prime}(\omega')}{\omega'-\omega}d\omega'\right] \\ &= -\frac{2\omega}{\pi}P.V.\int_{0}^{\infty} \frac{\chi^{(1)\prime}(\omega')}{(\omega'^2-\omega^2)}d\omega'.\end{aligned} \tag{2.A.9}$$

Thus we have proved the K–K relation expressions (2.3.4) and (2.3.5) in the linear polarization.

Appendix B: Two Systems of Units [11]

There are two different unit systems commonly used in nonlinear optics: one is the international system (System International, SI), or called the practical unit system to use the units of Meter, Kilogram and Second (i.e., MKS system); other one is the Gaussian system to use the units of centimeter, gram and second (i.e., cgs system). This unit system also can be called the electrostatic unit system (i.e., esu system). In short, there are two unit systems in nonlinear optics: International system (MKS/SI) and Gaussian system (cgs/esu). This book only uses the international unit system.

Here we briefly review these two unit systems and the conversion between two systems.

I. Fundamental Formula

Electric displacement formula:
MKS/SI unit system $\boldsymbol{D} = \varepsilon_0 \boldsymbol{E} + \boldsymbol{P}$
cgs/esu unit system $\boldsymbol{D} = \boldsymbol{E} + 4\pi \boldsymbol{P}$
Susceptibility formula:
MKS/SI unit system $\boldsymbol{P}(t) = \varepsilon_0[\boldsymbol{\chi}^{(1)}\boldsymbol{E}(t) + \boldsymbol{\chi}^{(2)}\boldsymbol{E}^2(t) + \boldsymbol{\chi}^{(3)}\boldsymbol{E}^3(t) + \cdots]$
cgs/es unit system $\boldsymbol{P}(t) = \boldsymbol{\chi}^{(1)}\boldsymbol{E}(t) + \boldsymbol{\chi}^{(2)}\boldsymbol{E}^2(t) + \boldsymbol{\chi}^{(3)}\boldsymbol{E}^3(t) + \cdots$

II. Conversion of Two Unit Systems

Electric field strength $\boldsymbol{E}(SI) = 3 \times 10^4 \boldsymbol{E}(esu)$
Linear susceptibility $\boldsymbol{\chi}^{(1)}(SI) = 4\pi\boldsymbol{\chi}^{(1)}(esu)$
Second-order susceptibility

$$\boldsymbol{\chi}^{(2)}(SI) = \frac{4\pi}{3 \times 10^4}\boldsymbol{\chi}^{(2)}(esu) = 4.189 \times 10^{-4}\boldsymbol{\chi}^{(2)}(esu)$$

Third-order susceptibility

$$\boldsymbol{\chi}^{(3)}(SI) = \frac{4\pi}{(3 \times 10^4)^2}\boldsymbol{\chi}^{(3)}(esu) = 1.40 \times 10^{-8}(esu)$$

n-order susceptibility

$$\boldsymbol{\chi}^{(n)}(SI) = \frac{4\pi}{(c \times 10^{-4})^{n-1}}\boldsymbol{\chi}^{(n)}(esu),\ \mathrm{c} = 3 \times 10^8$$

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