Chapter 3 General Description of Wave Propagation in a Nonlinear Medium

3.1 A General Form of the Problem

In this chapter, we are going to analyze the coupling effects between

(1) light waves, or occurring between

(2) light waves and the induced polarization.

To facilitate our discussion, let us assume an induced optical polarization to be the source term of the Maxwell's equations

$$\begin{aligned} \nabla\times E(r,t) &= -\frac{\partial B(r,t)}{\partial t} \\ \nabla\times B(r,t) &= \frac{4\pi}{c}J+\frac{1}{c}\frac{\partial E(r,t)}{\partial t} \end{aligned} \quad (1)$$

Here J denotes a total volume current density, which is composed of polarization current and conduction current such as

$$J=\frac{\partial P(r,t)}{\partial t}+J_c(r,t).$$

In NLO, we are usually not interested in the conduction current density $J_c(r,t)$. Therefore, let $J_c(r,t)=0$ for the time being. This is rigorously valid in a dielectric medium containing bound charges only.

Combining the two curl equations of Eq. (1) to form

$$\nabla\times\nabla\times E(\vec{r},t)+\frac{1}{c^2}\frac{\partial^2 E(\vec{r},t)}{\partial t^2}=-\frac{4\pi}{c^2}\frac{\partial^2 P(\vec{r},t)}{\partial t^2} \quad \text{[the time domain]} \quad (2)$$

and then transforming Eq. (2) into the frequency domain with $E(\vec{r},t)=\int E(\vec{r},\omega)e^{i\omega t}d\omega$,

we obtain

$$\nabla\times\nabla\times E(\vec{r},\omega)=\frac{\omega^2}{c^2}E(\vec{r},\omega)+\frac{4\pi\omega^2}{c^2}P(\vec{r},\omega) \quad \text{[the frequency domain]}. \quad (3)$$

3.1 Wave Propagation in the Linear Regime

In Eq. (3), let $\vec{P}(\vec{r},\omega)=\vec{P}_L(\vec{r},\omega)=\vec{\chi}^{(1)}(\omega)\cdot\vec{E}(\vec{r},\omega)$,

$$\nabla\times\nabla\times E(\vec{r},\omega)=\frac{\omega^2}{c^2}[\overleftrightarrow{I}+4\pi\overleftrightarrow{\chi}^{(1)}]\cdot\vec{E}(\vec{r},\omega)=\frac{\omega^2}{c^2}\overleftrightarrow{\varepsilon}(\omega)\cdot\vec{E}(\vec{r},\omega), \qquad (4)$$

where $\overleftrightarrow{\varepsilon}(\omega)=[\overleftrightarrow{I}+4\pi\,\overleftrightarrow{\chi}^{(1)}(\omega)]=\begin{bmatrix}1+4\pi\chi^{(1)}_{xx} & 4\pi\chi^{(1)}_{xy} & 4\pi\chi^{(1)}_{xz}\\ 4\pi\chi^{(1)}_{yx} & 1+4\pi\chi^{(1)}_{yy} & 4\pi\chi^{(1)}_{yz}\\ 4\pi\chi^{(1)}_{zx} & 4\pi\chi^{(1)}_{zy} & 1+4\pi\chi^{(1)}_{zz}\end{bmatrix}$.

Here we consider a wave propagating in a linear ($P_{NL}=0$) while anisotropic ($\varepsilon_{ij}\neq 0$) medium. The governing wave equation becomes

$$\nabla\times[\nabla\times E(\vec{r},\omega)]-\frac{\omega^2}{c^2}\vec{E}(\vec{r},\omega)=\frac{4\pi\omega^2}{c^2}\overleftrightarrow{\chi}^{(1)}(\omega)\cdot\vec{E}(\vec{r},\omega) \quad .$$

By combining the last two terms together, we then obtain

$$\nabla\times[\nabla\times E(\vec{r},\omega)]-\frac{\omega^2}{c^2}\overleftrightarrow{\varepsilon}\cdot\vec{E}(\vec{r},\omega)=0 \quad .$$
$$\textit{with}\quad \overleftrightarrow{\varepsilon}(\omega)=\overleftrightarrow{I}+4\pi\,\overleftrightarrow{\chi}^{(1)}(\omega)$$

First, let us consider a monochromatic wave propagating in a uniform dielectric medium, the above wave equation can be simplified into

$$\nabla^2\vec{E}(\vec{r},\omega)+k_0^{\,2}n^2\vec{E}(\vec{r},\omega)=0\,,$$

where $k_0=\frac{\omega}{c}=\frac{2\pi}{\lambda_0}$ with k_0=wave vector in vacuum, and n=the refractive index of the medium. When the wave propagates in presence of fluctuations n(x, y, z; t), a term coupling the polarizations may occur in the wave equation

$$\nabla^2\vec{E}(\vec{r},\omega)+k_0^{\,2}n^2(\vec{r})\vec{E}(\vec{r},\omega)-2\nabla\left(\frac{\nabla n(\vec{r})}{n(\vec{r})}\cdot\vec{E}(\vec{r},\omega)\right)=0\,.$$

However, the order of magnitude calculations indicates that the coupling term may be negligible in an approximation and therefore, the fluctuations in refractive index do not cause mixing in the polarization components during the propagation. That is: **turbulent propagation still satisfies the "scalar diffraction" picture.**

In this case, we can decompose $n(\vec{r})\ =\ <n>+\delta n(\vec{r})=n_0+\delta n(\vec{r})$, and let $k=k_0n_0$ to denote the average wave vector in the unperturbed medium. Then,

$$\nabla^2\vec{E}(\vec{r},\omega)+k^2\,(1+2\frac{\delta n(\vec{r})}{n_0})\,\vec{E}(\vec{r},\omega)=0\,.$$

Thus, the third term can be considered as a perturbation to

$$\nabla^2\vec{E}(\vec{r},\omega)+k_0^{\,2}n^2\vec{E}(\vec{r},\omega)=0\,.$$

.

Now let us return to the discussion on the uniform while anisotropic dielectric medium. We can find a set of possible solution of Eq. (4) to be

$$\vec{E}(\vec{r},\omega)=\vec{E}_0(\omega)e^{i\vec{K}\cdot\vec{r}}, \quad (5)$$

where

$\vec{K}=\frac{\omega}{c}[n_r(\omega)+in_i(\omega)]\hat{s}=\tilde{\omega}\,n\,\hat{s}$, $\vec{E}_0(\omega)=\hat{e}\,E_0(\omega)$ by defining $\tilde{\omega}=\omega/c\,,\vec{k}=\tilde{\omega}n\,\hat{s}$.

By plugging the possible solution set of Eq. (5) into $\nabla\times[\nabla\times E(\vec{r},\omega)]-\frac{\omega^2}{c^2}\vec{\varepsilon}\cdot\vec{E}(\vec{r},\omega)=0$, we obtain

$$\vec{k}\times[\vec{k}\times E(\vec{r},\omega)]+\frac{\omega^2}{c^2}\vec{\varepsilon}\cdot\vec{E}(\vec{r},\omega)=0\,. \quad (6)$$

Note $\vec{\chi}^{(1)}$ is a symmetric 2nd-rank tensor $\chi_{ij}^{(1)}=\chi_{ji}^{(1)}$. Since $\varepsilon_{ij}=\varepsilon_{ji}$, we can diagonalize $\vec{\varepsilon}$ and express Eq. (6) in terms of the principal axes of $\vec{\varepsilon}$ as

$$\begin{bmatrix}\tilde{\omega}^2\varepsilon_x-k_y^2-k_z^2 & k_xk_y & k_xk_z\\ k_yk_x & \tilde{\omega}^2\varepsilon_y-k_x^2-k_z^2 & k_yk_z\\ k_zk_x & k_zk_y & \tilde{\omega}^2\varepsilon_z-k_x^2-k_y^2\end{bmatrix}\begin{pmatrix}E_x\\E_y\\E_z\end{pmatrix}=0\,. \quad (7)$$

To have a nontrivial solution of $\vec{E}$, the determinant of the Eq. (7) must be equal to zero

$$[\textbf{determinant}]=0\,. \quad (8)$$

Eq. (8) can lead to the following interesting results:

- For a given propagation direction $\hat{s}$, there are two k values which are the intersections of the direction of propagation and the normal surface. Thus, we have two different phase velocities $\omega/k_{1,2}$ in the medium. Let $|\vec{k}|=\tilde{\omega}n$, the fact of two values of $|\vec{k}|$ implies there are two different indices of refraction.
- The directions of the electric field vector $\vec{E}_0(\omega)=\hat{e}\,E_0(\omega)$ associated with these propagations can be given by $\hat{e}_{1,2}=\begin{pmatrix}k_x/(k_{1,2}{}^2-\tilde{\omega}^2\varepsilon_x)\\ k_y/(k_{1,2}{}^2-\tilde{\omega}^2\varepsilon_y)\\ k_z/(k_{1,2}{}^2-\tilde{\omega}^2\varepsilon_z)\end{pmatrix}$.
- The two phase velocities correspond to two mutually orthogonal polarizations.

Let $\vec{k}=\tilde{\omega}n\,\hat{s}$, Eq. (8) reduces to the well-known Fresnel's equation of wave normal

$$\frac{s_x^2}{n^2-\varepsilon_x}+\frac{s_y^2}{n^2-\varepsilon_y}+\frac{s_z^2}{n^2-\varepsilon_z}=\frac{1}{n^2} \qquad (9)$$

and the directions of the electric field vector become $\hat{e}_{1,2}=\begin{pmatrix} s_x/(n_{1,2}^2-\varepsilon_x) \\ s_y/(n_{1,2}^2-\varepsilon_y) \\ s_z/(n_{1,2}^2-\varepsilon_z) \end{pmatrix}$.

Eq. (9) is a quadratic equation of n^2. Note that in a dielectric medium without free charges $\nabla\cdot D=0$. The independent components of $\vec{D}$ is two, thus we can choose $\vec{D}_1\cdot\vec{D}_2$ =0, and form an orthogonal triad $\{\vec{D}_1,\vec{D}_2,\hat{s}\}$ for a given propagation direction $\hat{s}$. By using these notations and from

$$\nabla\times B=\frac{1}{c}\frac{\partial D}{\partial t} \qquad \textbf{(a)}$$

$$\nabla\times E=-\frac{1}{c}\frac{\partial B}{\partial t} \qquad \textbf{(b)}$$

with $B=\mu H$. In terms of the plane-wave solution set, Eqs. (a&b) can be rewritten as

$$-i\,\tilde{\omega}\vec{D}=-i\,\tilde{\omega}n\,\mu\,\hat{s}\times\vec{H} \qquad \textbf{(a)}$$

$$-i\,\tilde{\omega}\mu\vec{H}=-i\,\vec{k}\times\vec{E}=-i\,\tilde{\omega}n\,\hat{s}\times\vec{E} \qquad \textbf{(b)}.$$

Poynting vector can be used to reveal the energy flow of an optical beam in a medium, thus

$$\vec{P}=\textbf{Poynting Vector }=\vec{E}\times\vec{H}=\frac{n}{\mu}\vec{E}\times(\hat{s}\times\vec{E})=\frac{n}{\mu}\{\hat{s}\,|\,E\,|^2-(\vec{E}\cdot\hat{s})\vec{E}\}$$

$$=\textbf{is not parallel to }\hat{s}.$$

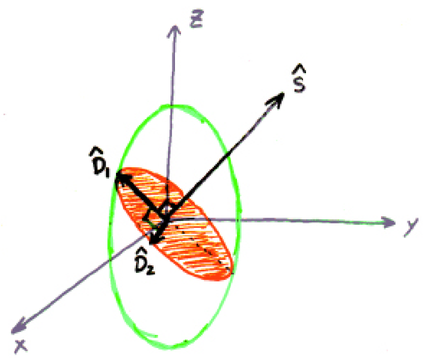
We can also estimate the electric energy stored in the medium to be

$$U_e=\textbf{Electric Energy }=\frac{1}{2}\vec{E}\cdot\vec{D}$$

$$\Rightarrow\quad \frac{D_x^2}{\varepsilon_x}+\frac{D_y^2}{\varepsilon_y}+\frac{D_z^2}{\varepsilon_z}=2U_e \qquad \textbf{(10)}.$$

Here $\varepsilon_x,\varepsilon_y,\varepsilon_z$ denote the principal dielectric constants.

By defining $D_x/\sqrt{2U_e}=x$, Eq. (10) then becomes $\frac{x^2}{n_x^{\ 2}}+\frac{y^2}{n_y^{\ 2}}+\frac{z^2}{n_z^{\ 2}}=1$, which is the **Index Ellipsoid** of a medium. Let $n_x=n_y$, the ellipsoid reduces to a rotational ellipsoid along the *z*-axis:



- We can define an impermeability tensor η as $\eta_{ij} = [1/\varepsilon]_{ij}$ $\Rightarrow$ $\vec{E} = \vec{\eta} \cdot \vec{D}$.

Thus, Eq. (6) $\vec{k} \times (\vec{k} \times \vec{E}) + \tilde{\omega}^2 \vec{\varepsilon} \cdot \vec{E} = 0$ becomes

$$(n\tilde{\omega})^2 \hat{s} \times (\hat{s} \times \vec{E}) + \tilde{\omega}^2 \vec{\varepsilon} \cdot \vec{E} = 0 \quad \Rightarrow \quad \hat{s} \times [\hat{s} \times (\vec{\eta} \cdot \vec{D}) + \frac{1}{n^2} \vec{D} = 0.$$

Redefine a new coordinate system such that one of the coordinate axes along the beam propagation direction $\hat{s}$, and let $\hat{s} = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}$, then

$$\hat{s} \times [\hat{s} \times (\vec{\eta} \cdot \vec{D}) = \hat{s}(\vec{\eta} \cdot \vec{D})_z - (\vec{\eta} \cdot \vec{D}) = -(\vec{\eta} \cdot \vec{D})_T$$

$$\Rightarrow \quad (\eta_T - \frac{1}{n^2}) D = 0.$$

This implies two other axes shall be along the eigenvectors of η_T : $\{ \hat{D}_1, \hat{D}_2 \}$.

The inverse index of refraction associated with any given propagation direction $\hat{s}$ can then be expressed as a linear combination of two transverse eigenvectors

$$\frac{1}{n_e^2(\theta)} = \frac{\cos^2\theta}{n_O^2} + \frac{\sin^2\theta}{n_E^2}.$$

3.3 Wave Propagation in a Nonlinear Medium ($P_{NL} \neq 0$)

From the wave equation with an induced polarization $P(\vec{k}, \omega)$

$$\nabla \times [\nabla \times E(\vec{k}, \omega)] = \frac{\omega^2}{c^2} \vec{E}(\vec{k}, \omega) + \frac{4\pi\omega^2}{c^2} P(\omega, \vec{k}_m)$$

By separating $P(\vec{k}_m, \omega) = P_L(\vec{k}_m, \omega) + P_{NL}(\vec{k}_m, \omega)$ and define $\overleftrightarrow{\varepsilon}(\omega) = \overleftrightarrow{I} + 4\pi\overleftrightarrow{\chi}^{(1)}(\omega)$, we then obtain

$$\nabla\times[\nabla\times E(\vec{k},\omega)] = \frac{\omega^2}{c^2}\overleftrightarrow{\varepsilon}\cdot\vec{E}(\vec{k},\omega) + \frac{4\pi\omega^2}{c^2}P_{NL}(\vec{k}_m,\omega_m=\omega) \quad . \tag{1}$$

Here $P_{NL}(\vec{k}_m,\omega_m=\omega)$ is a nonlinear polarization which can be generated from a generalized driving force with the product of $E_1(\vec{k}_1,\omega_1)],\cdots,E_n(\vec{k}_n,\omega_n)]$.

Now note that

- ω_m shall be equal to ω in $P_{NL}(\vec{k}_m,\omega_m)$ because of photon energy must be conserved in the steady-state case.
- But $\vec{k}_m$ needs not be exactly equal to $\vec{k}$. This is because wave momentum conservation is not strictly required in a finite-sized medium with dimensions comparable to the optical wavelengths involved.
- Eq. (1) forms a set of (n+1) coupled wave equations (coupled together through nonlinear polarization $P_{NL}(\vec{k}_m,\omega_m)$).

Let us examine the equation with second-order nonlinear optical effect:

$\vec{E}_1(\vec{k}_1,\omega_1), \vec{E}_2(\vec{k}_2,\omega_2) \rightarrow \vec{E}_s(\vec{k}_s,\omega_s=\omega_1+\omega_2)$,

the governing equations become

$$\begin{cases} [\nabla\times\nabla\times - \frac{\omega_1^{\ 2}}{c^2}\overleftrightarrow{\varepsilon}(\omega_1)\cdot\,]\,\vec{E}_1(\vec{k}_1,\omega_1) = \frac{4\pi\omega_1^{\ 2}}{c^2}\chi^{(2)}(\omega_1=\omega_s-\omega_2):\vec{E}_2^{\ *}(\vec{k}_2,\omega_2)\vec{E}_s(\vec{k}_s,\omega_s) \\ [\nabla\times\nabla\times - \frac{\omega_2^{\ 2}}{c^2}\overleftrightarrow{\varepsilon}(\omega_2)\cdot\,]\,\vec{E}_2(\vec{k}_2,\omega_2) = \frac{4\pi\omega_2^{\ 2}}{c^2}\chi^{(2)}(\omega_2=\omega_s-\omega_1):\vec{E}_s(\vec{k}_s,\omega_s)\vec{E}_1^{\ *}(\vec{k}_1,\omega_1)\cdot \\ [\nabla\times\nabla\times - \frac{\omega_s^{\ 2}}{c^2}\overleftrightarrow{\varepsilon}(\omega_s)\cdot\,]\,\vec{E}_s(\vec{k}_s,\omega_s) = \frac{4\pi\omega_s^{\ 2}}{c^2}\chi^{(2)}(\omega_s=\omega_1+\omega_2):\vec{E}_1(\vec{k}_1,\omega_1)\vec{E}_2(\vec{k}_2,\omega_2) \end{cases}$$

3.4 Slowly Varying Amplitude Approximation (SVA)

In this section, we will consider the wave couplings in a NLO medium, leading to

- an energy transfer among waves, and causes
- optical field amplitudes to change with wave propagation.

Considering a plane wave propagating along z, $E(\omega,\vec{r},t)=\mathcal{E}(\rho,z,t)e^{i(kz-\omega t)}$, the energy transfer among waves is significant only after the waves travel over a distance which is much longer than their wavelengths, that is,

$$\left|\frac{\partial\mathcal{E}}{\partial z}\right|\gg\frac{\lambda}{2\pi}\left|\frac{\partial^2\mathcal{E}(z)}{\partial z^2}\right| \quad i.e., \quad \left|\frac{\partial^2\mathcal{E}(z)}{\partial z^2}\right|\Big/\left|\frac{\partial\mathcal{E}}{\partial z}\right|\sim\delta k\ll k\ .$$

Based on the radiation theory, we can split the nonlinear wave propagation equation $\nabla\times[\nabla\times E(\vec{r},\vec{k},\omega)]+\frac{\omega^2}{c^2}\vec{\varepsilon}(\omega)\cdot\vec{E}(\vec{r},\vec{k},\omega)=-\frac{4\pi\omega^2}{c^2}P_{NL}(\vec{r},\omega,\vec{k}_m)$ into two parts according to the source is perpendicular or parallel to the wave propagation direction:

$$\begin{aligned}&\nabla^2E_\perp+\frac{\omega^2}{c^2}[\vec{\varepsilon}\cdot\vec{E}]_\perp=-\frac{4\pi\omega^2}{c^2}P_{NL,\perp}\\&\nabla\cdot[(\vec{\varepsilon}\cdot\vec{E})_{\|}+4\pi P_{NL,\|}]=0\end{aligned}.$$

Let $E_\perp(\vec{r},t)=\mathcal{E}(\rho,z,t)$, then $\nabla^2E_\perp=(\nabla_\perp{}^2+\frac{\partial^2}{\partial z^2})\mathcal{E}(\rho,z,t)$. Neglecting the diffraction effect of the first term,

$$\begin{aligned}\nabla^2E_\perp&=\frac{\partial^2}{\partial z^2}\mathcal{E}(\rho,z,t)=\frac{\partial^2}{\partial z^2}[\mathcal{E}_\perp(\rho,z)e^{i(kz-\omega t)}]\\&=e^{i(kz-\omega t)}[\frac{\partial^2\mathcal{E}_\perp}{\partial z^2}+2ik\frac{\partial\mathcal{E}_\perp}{\partial z}-k^2]\end{aligned}.$$

Now by invoking SVA, the coupled wave equation $\nabla^2E_\perp+\frac{\omega^2}{c^2}[\vec{\varepsilon}\cdot\vec{E}]_\perp=-\frac{4\pi\omega^2}{c^2}P_{NL,\perp}$ becomes

$$\frac{\partial\mathcal{E}_\perp(\omega,z)}{\partial z}=\frac{2\pi i\omega^2}{kc^2}P_{NL,\perp}(\omega,z)e^{-i(kz-\omega t)}.$$

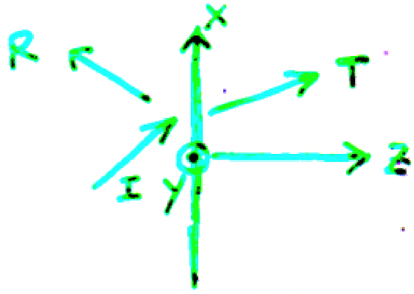
For simplicity, we will focus on the planar wave propagation without considering the diffraction effect. Therefore, the field dependence on transverse coordinates ρ will be neglected hereafter.

By writing $P_{NL,\perp}(\omega_s,z)=P_\perp^{NL}(z)e^{i(k_sz-\omega_st)}$,

$$\frac{\partial\mathcal{E}_\perp}{\partial z}=\frac{2\pi i\omega^2}{kc^2}P_\perp^{NL}(z)e^{-i\Delta kz}\quad \text{with}\quad \Delta k=k-k_s\ .$$

To solve the coupled equations, we need to implement some **boundary conditions**, which are

- Tangential components of $\vec{E}$ **and** $\vec{B}$ at a boundary surface must be continuous for each Fourier component. Since $\vec{E}=\vec{E}_H+\vec{E}_P=$ homogeneous + particular solutions, the boundary conditions imply



$$\begin{cases}(\vec{E}_R+\vec{E}_I)_x=(\vec{E}_T)_x\\(\vec{k}_I\times\vec{E}_I)_x+(\vec{k}_R\times\vec{E}_R)_x=(\vec{k}_T\times\vec{E}_T)_x\\(\vec{k}_I\times\vec{E}_I)_y+(\vec{k}_R\times\vec{E}_R)_y=(\vec{k}_T\times\vec{E}_T)_y\end{cases}.$$

Due to the translation symmetry on the boundary,

$$\begin{cases}k_{I,x}(\text{homogeneous})=k_{I,x}(\text{particular})\\=k_{R,x}(\text{homogeneous})=k_{R,x}(\text{particular})\\=k_{T,x}(\text{homogeneous})=k_{T,x}(\text{particular})\end{cases}.$$

3.5 Time-Dependent Wave Propagation

Waves with time-varying amplitude should obey the wave equation in time-domain

$$[\nabla\times(\nabla\times)+\frac{1}{c^2}\frac{\partial^2}{\partial t^2}]\vec{E}(\vec{r},t)=-\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\vec{P}(\vec{r},t).$$

Assuming that a quasi-monochromatic plane wave propagates along the symmetry axis $\hat{z}$, then

$$\frac{\partial^2\vec{E}}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2\vec{D}}{\partial t^2}=\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\vec{P}_{NL}(z,t),$$

where $\vec{D}(z,t)=\vec{E}(z,t)+4\pi\vec{P}_L(z,t)$.

Let $\vec{E}(z,t)=\vec{\mathcal{E}}(z,t)e^{ikz-i\omega t}$, then

$$\frac{\partial^2\vec{E}(z,t)}{\partial z^2}\simeq(2ik\frac{\partial}{\partial z}\vec{\mathcal{E}}-k^2\vec{\mathcal{E}})e^{ikz-i\omega t}.$$

Here we have applied SVA on this equation.

By expressing $\vec{E}(z,t)$ in terms of Fourier integral $E(z,t)=\int\mathcal{E}(\omega+\eta)e^{ikz-i(\omega+\eta)t}d\eta$,

therefore $D(z,t)=\int\varepsilon(\omega+\eta)\mathcal{E}(\omega+\eta)e^{ikz-i(\omega+\eta)t}d\eta$.

We examine $\frac{1}{c^2}\frac{\partial^2\vec{D}(z,t)}{\partial t^2}$ in more detail in terms of Fourier integral

$$\frac{1}{c^2}\frac{\partial^2 D(z,t)}{\partial t^2}=\int -\frac{(\omega+\eta)^2}{c^2}\varepsilon(\omega+\eta)\mathcal{E}(\omega+\eta)e^{ikz-i(\omega+\eta)t}$$

$$=-\int[\frac{\omega^2\varepsilon(\omega)}{c^2}+\frac{\omega^2\eta}{c^2}\frac{\partial\varepsilon}{\partial\omega}|_\omega+\frac{2\omega\eta\varepsilon(\omega)}{c^2}+\mathcal{O}(\eta^2)]\mathcal{E}(\omega+\eta)e^{ikz-i(\omega+\eta)t}d\eta$$

$$\simeq -\frac{\omega^2}{c^2}\int\varepsilon(\omega)\mathcal{E}(\omega+\eta)e^{ikz-i(\omega+\eta)t}d\eta-\frac{\omega^2}{c^2}\int\eta\frac{\partial\varepsilon}{\partial\omega}|_\omega\cdot\mathcal{E}(\omega+\eta)e^{ikz-i(\omega+\eta)t}d\eta$$

$$-\frac{2\omega}{c^2}\int\eta\cdot\varepsilon(\omega)\cdot\mathcal{E}(\omega+\eta)e^{ikz-i(\omega+\eta)t}d\eta$$

We can calculate the group velocity of an optical pulse in a dispersive medium with

$$\frac{1}{v_g}\equiv\frac{dk}{d\omega}=\frac{d}{d\omega}(\frac{\sqrt{\varepsilon(\omega)}\ \omega}{c})=\frac{\sqrt{\varepsilon(\omega)}}{c}+\frac{\omega}{2c\sqrt{\varepsilon(\omega)}}\frac{d\varepsilon(\omega)}{d\omega},\ i.e.,\ \ \frac{d\varepsilon(\omega)}{d\omega}=\frac{2c\sqrt{\varepsilon}}{\omega}\frac{1}{v_g}-\frac{2\varepsilon}{\omega}.$$

Thus, we obtain

$$2\omega\eta\varepsilon+\omega^2\eta\frac{\partial\varepsilon}{\partial\omega}=\frac{2kc^2}{v_g}\eta \quad \text{and}$$

$$\int\eta\mathcal{E}(\omega+\eta)e^{ikz-i(\omega+\eta)t}dt=i\frac{\partial}{\partial t}\mathcal{E}(z,t)$$

We can reduce the term of $\frac{1}{c^2}\frac{\partial^2\vec{D}(z,t)}{\partial t^2}$ in the time-dependent wave equation into

$$\frac{1}{c^2}\frac{\partial^2 D(z,t)}{\partial t^2}=[-\frac{\omega^2}{c^2}\varepsilon(\omega)\mathcal{E}(z,t)-\frac{2ik}{v_g}\frac{\partial\mathcal{E}(z,t)}{\partial t}]e^{ikz-i\omega t}.$$

We then approximate $\frac{\partial^2 P_{NL}(z,t)}{\partial t^2}\simeq-\omega^2 P_{NL}(z,t)$ and finally obtain

$$(\frac{\partial}{\partial z}+\frac{1}{v_g}\frac{\partial}{\partial t})\mathcal{E}_{\mathcal{F}}(z,t)=\frac{2\pi i\omega^2}{kc^2}P_{NL}(z,t)e^{i(kz-\omega t)} \quad \textbf{[Forward Propagation]}$$

to depict a wave propagating in the +*z* direction.
Similarly,

$$(\frac{\partial}{\partial z}-\frac{1}{v_g}\frac{\partial}{\partial t})\mathcal{E}_{\mathcal{B}}(z,t)=-\frac{2\pi i\omega^2}{kc^2}P_{NL}(z,t)e^{i(kz+\omega t)} \quad \textbf{[Backward Propagation]}$$

to depict a wave propagating in the -*z* direction.

If $\left|[l\sqrt{\varepsilon(\omega)}/c]\cdot(\partial\mathcal{E}/\partial t)\right|\ll|\mathcal{E}|$, and note $|l\sqrt{\varepsilon(\omega)}/c|=t_{tr}\cdot$ **and** $|(\partial\mathcal{E}/\partial t)|=|\mathcal{E}|/t_p$ the inequality leads to $t_{tr}\ll t_p$. Under this condition, the time derivative term can be neglected and this renders into the wave propagation in the steady-state regime.

3.6 The Relationship between Macroscopic and Local Field Quantities

Note that

- Fields appearing in the Maxwell's equations are macroscopic quantities, *i.e.*, they are averaged over macroscopic volume of polarization.

- But dynamical model of polarization $\vec{P}(\vec{r})$ often requires one to take into account local field at the position of a particular molecule.

To solve the difficulty, let us first consider the following situation:

♦ **a linear, isotropic medium** (*i.e.*, fluid or crystal with cubic symmetry) $P_{NL} = 0$

$$E_{loc} = E_{ext} + \frac{4\pi}{3}P_L = E_{ext} + \frac{4\pi}{3}\chi^{(1)}E_{loc}$$
$$\Rightarrow \quad E_{loc} = \frac{E_{ext}}{1 - \frac{4\pi}{3}\chi^{(1)}} \simeq [1 + \frac{4\pi}{3}\chi^{(1)}]E_{ext} \qquad .$$
$$\Rightarrow \quad E_{loc} = \frac{2 + 1 + 4\pi\chi^{(1)}}{3}E_{ext} = [\frac{2+\varepsilon(\omega)}{3}]E_{ext} \equiv L(\omega)E_{ext}$$

Then consider

♦ **a nonlinear medium** with $\chi^{(2)} \neq 0$ (*i.e.*, $P_{NL} \neq 0$)

$$E_{loc} = E_{ext} + \frac{4\pi}{3}(P_L + P_{NL}),$$

where $P_L = \chi^{(1)}E_{loc}$ denotes the linear polarization, and P_{NL} indicates the nonlinear polarization.

Thus,

$$E_{loc} = E_{ext} + \frac{4\pi}{3}\chi^{(1)}E_{loc} + \frac{4\pi}{3}P_{NL}$$
$$\Rightarrow \quad E_{loc} = \frac{(E_{ext} + \frac{4\pi}{3}P_{NL})}{(1 - \frac{4\pi}{3}\chi^{(1)})} \quad ,$$

implying the linear dipole moment can be affected by the existing nonlinear dipole moments in the neighborhood by

$$P_L = \chi^{(1)} E_{loc} = \frac{\chi^{(1)}}{1-\frac{4\pi}{3}\chi^{(1)}} \cdot (E_{ext} + \frac{4\pi}{3} P_{NL}) .$$

Note that by using Clausius-Mossotti equation $\chi^{(1)} = \frac{3}{4\pi}\frac{\varepsilon-1}{\varepsilon+2}$, we can further convert linear polarization to be

$$P_L = \frac{\varepsilon-1}{4\pi} E_{ext} + \frac{\varepsilon-1}{3} P_{NL} .$$

Recall

$$\begin{aligned} D &\equiv E_{ext} + 4\pi P = E_{ext} + 4\pi P_L + 4\pi P_{NL} \\ &= E_{ext} + (\varepsilon-1)E_{ext} + \frac{4\pi}{3}(\varepsilon-1)P_{NL} + 4\pi P_{NL} \\ &= \varepsilon E_{ext} + 4\pi(\frac{\varepsilon+2}{3})P_{NL} \\ &= \varepsilon E_{ext} + 4\pi P_{NL,\,eff} \end{aligned} .$$

Assuming the nonlinear optical polarization to be second-order effect $P_{NL} = \chi^{(2)} E_{loc} E_{loc}$, then

$$\begin{aligned} P_{NL,eff} &\equiv \chi^{(2)}_{eff} : E_{ext} E_{ext} = \frac{\varepsilon+2}{3} P_{NL} \\ &\equiv \frac{\varepsilon(\omega_3)+2}{3} \cdot \chi^{(2)}_{loc}(-\omega_3;\omega_2,\omega_1) E_{loc}(\omega_2) E_{loc}(\omega_1) \\ &= [\frac{\varepsilon(\omega_3)+2}{3}] \chi^{(2)}_{loc}(-\omega_3;\omega_2,\omega_1) [\frac{\varepsilon(\omega_2)+2}{3}][\frac{\varepsilon(\omega_1)+2}{3}] E_{ext}(\omega_2) E_{ext}(\omega_1) \end{aligned} .$$

This implies that we can define an effective nonlinear susceptibility with

$$\begin{aligned} \chi^{(2)}_{eff,ijk}(-\omega_3;\omega_2,\omega_1) &= [\frac{\varepsilon(\omega_3)+2}{3}]_{i\alpha} [\frac{\varepsilon(\omega_2)+2}{3}]_{j\beta} [\frac{\varepsilon(\omega_1)+2}{3}]_{k\gamma} \chi^{(2)}_{loc,\alpha\beta\gamma}(-\omega_3;\omega_2,\omega_1) \\ &= L_{i\alpha}(\omega_3) L_{j\beta}(\omega_2) L_{k\gamma}(\omega_1) \chi^{(2)}_{loc,\alpha\beta\gamma}(-\omega_3;\omega_2,\omega_1) \end{aligned} .$$

However we shall keep in mind that the results have neglected the interaction between nonlinear dipole moments at different sites.