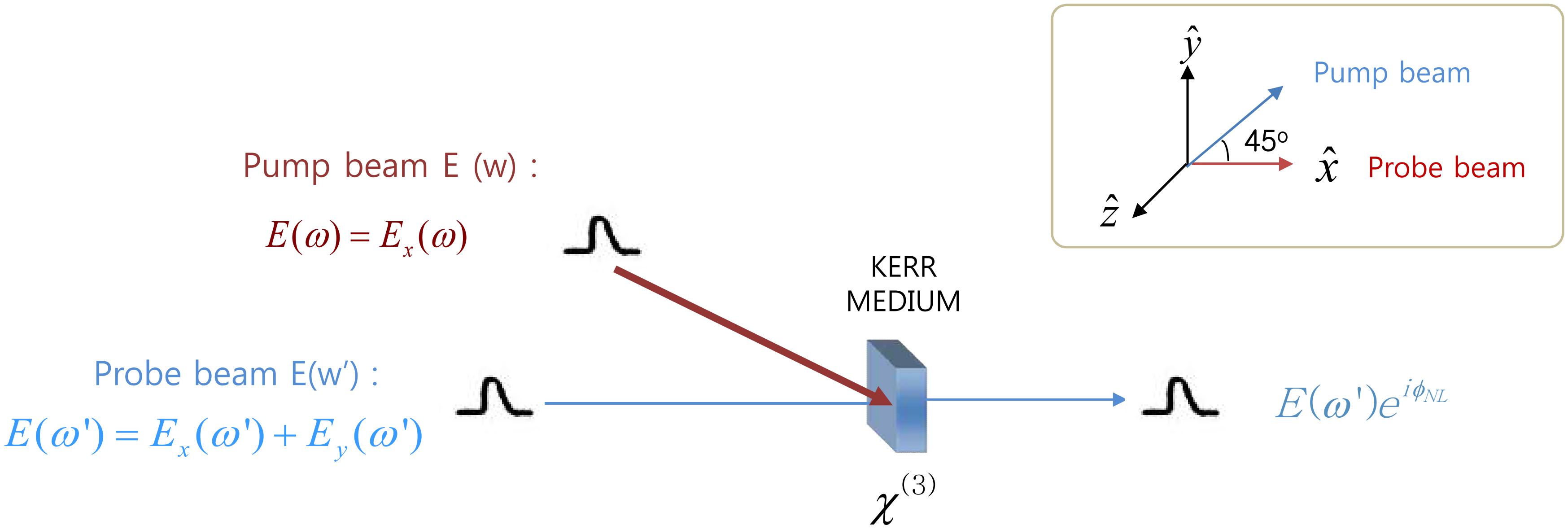
Optical Kerr Effect Theory

2010.8.21 Jun heesun

Optical kerr effect

Nonlinear phase shift induced by the strong pump beam is picked up by the probe beam passing through crossed polarizes.



Optical kerr effect polarization in an isotropic medium

Pump beam E (w) : $E(\omega) = E_x(\omega)$

Probe beam E(w') : $E(\omega') = E_x(\omega') + E_y(\omega')$

$$P_i^{(3)}(\omega') = \sum_{j,k,l} \chi_{ijkl}^{(3)}(\omega' = \omega' + \omega - \omega) E_j(\omega') E_k(\omega) E_l(\omega)$$

$$\begin{aligned} P_x^{(3)}(\omega') &= \chi_{1111}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_1(\omega) E^*{}_1(\omega) \\ &\quad + \cancel{\chi_{1211}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_1(\omega) E^*{}_1(\omega)} \qquad (\because \text{isotropic medium}) \\ &= (\chi_{1122}^{(3)} + \chi_{1212}^{(3)} + \chi_{1221}^{(3)}) E_x(\omega') E(\omega) E^*(\omega) \end{aligned}$$

$$\begin{aligned} P_y^{(3)}(\omega') &= \cancel{\chi_{2111}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_1(\omega) E_1^*(\omega)} \\ &\quad + \chi_{2211}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_1(\omega) E_1^*(\omega) \qquad (\because \text{isotropic medium}) \\ &= \chi_{1122}^{(3)}(\omega' = \omega' + \omega - \omega) E_y(\omega') E(\omega) E^*(\omega) \qquad (\because \chi_{2211} = \chi_{1122}) \end{aligned}$$

The field-induced anisotropy in the susceptibility

$$\delta\chi\ (\omega') = \Delta\chi_{xx} - \Delta\chi_{yy} = (\chi_{1212} + \chi_{1221})\left|E_0(\omega)\right|^2$$

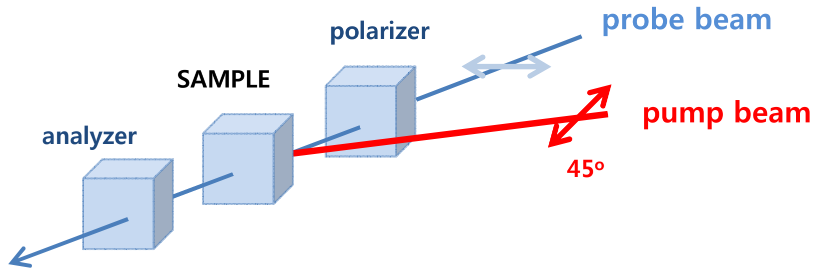
The induced linear birefringence given by

$$\delta n(\omega') = \frac{2\pi}{n_0}\delta\chi(\omega') = \frac{2\pi}{n_0}\left(\chi_{1212}^{(3)} + \chi_{1221}^{(3)}\right)\left|E_0(\omega)\right|^2$$

In propagating through a medium of length L, the $\hat{x}$ and $\hat{y}$ components of the probe field experience a relative phase difference

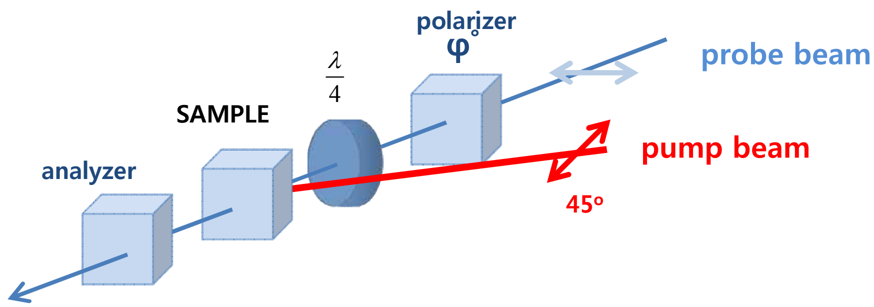
$$\delta\phi = (\frac{n_{xx}\omega}{c}L - \omega t) - (\frac{n_{yy}\omega}{c}L - \omega t) = \left(\frac{\omega}{c}\right)\delta n L$$

✓ **homodyne method optical Kerr Effect signal :**

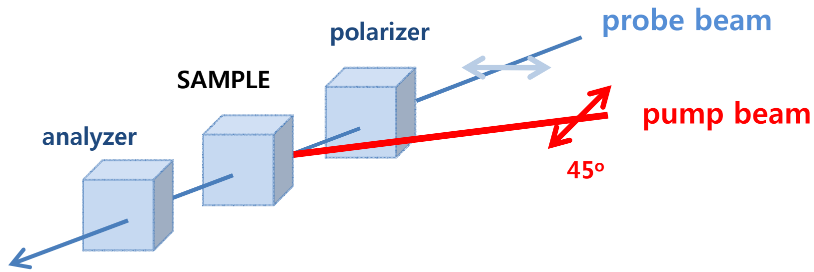


✓ **OHD-OKE(Optical Heterodyne Detected-Optical Kerr Effect) signal :**

$$S_{OHD\text{-}OKE}(t) = I_{background} + I_{homodyne} + I_{heterodyne}$$



✓ **homodyne method optical Kerr Effect signal Jones Matrix :**

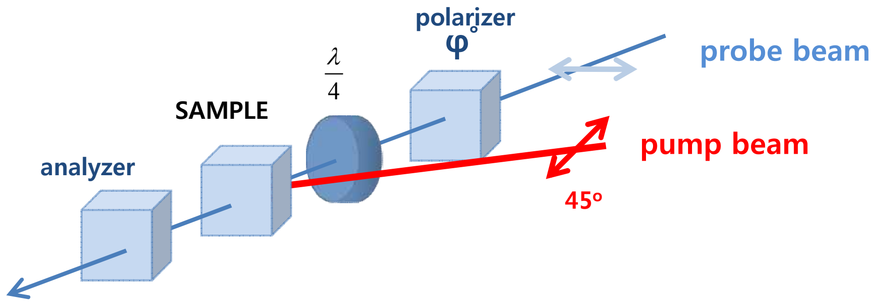


$$I = \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} \cos 45^o & -\sin 45^o \\ \sin 45^o & \cos 45^o \end{bmatrix} \begin{bmatrix} e^{i\phi_x} & 0 \\ 0 & e^{i\phi_y} \end{bmatrix} \begin{bmatrix} \cos 45^o & \sin 45^o \\ -\sin 45^o & \cos 45^o \end{bmatrix} \begin{bmatrix} 1 \\ 0 \end{bmatrix} = \sin^2[\frac{\phi_x}{2} - \frac{\phi_y}{2}]$$

analyzer — SAMPLE — probe beam

$$I =< E^2 >= \frac{\Delta\phi^2}{4}$$

✓ OHD-OKE Jones Matrix



$$I = \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} \cos 45^o & -\sin 45^o \\ \sin 45^o & \cos 45^o \end{bmatrix} \begin{bmatrix} e^{i\phi_x} & 0 \\ 0 & e^{i\phi_y} \end{bmatrix} \begin{bmatrix} \cos 45^o & \sin 45^o \\ -\sin 45^o & \cos 45^o \end{bmatrix} \begin{bmatrix} 1 & 0 \\ 0 & i \end{bmatrix} \begin{bmatrix} \cos\varphi & -\sin\varphi \\ \sin\varphi & \cos\varphi \end{bmatrix} \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} \cos\varphi & \sin\varphi \\ -\sin\varphi & \cos\varphi \end{bmatrix} \begin{bmatrix} 1 \\ 0 \end{bmatrix} = \cos[\varphi]^2 \sin[\frac{1}{2}(2\varphi + \phi_x - \phi_y)]^2$$

analyzer | SAMPLE | $\frac{\lambda}{4}$ | polarizer φ° | probe beam

$$I =< E^2 >= \varphi^2 + \varphi\Delta\phi + \frac{\Delta\phi^2}{4}$$

The effects of inertial motion were invoked to describe early depolarized light scattering spectra [9], [32] and the spectra of filaments formed from the self-focusing of intense laser pulses [10] in liquid CS_2. These workers treated the orientation of the condensed phase molecules with respect to the polarized radiation field as a linear driven oscillator described by the equation of motion:

$$I \frac{d^2\delta\theta}{dt^2} + \xi \frac{d\delta\theta}{dt} + \mu\delta\theta = F(t) \tag{7}$$

where $\delta\theta$ is the angle induced between the laser electric field and the principle axis of the molecular polarizability tensor, I is the molecular moment of inertia, ξ is the microscopic friction, μ is the elastic force constant of an harmonic potential, and $F(t)$ is the generalized forcing function. The form of the observed response function $\delta\theta(t)$ depends upon the relative magnitudes of the coefficients of (7) as well as the details of the forcing function, and has been evaluated by the Green's function method [33].

The equation of motion for the birefringence induced by an arbitrarily shaped laser pulse I_{pump} was derived from (7) and applied to filament spectra [10] in CS_2 in the form

$$I/\mu \frac{d^2\Delta n_i}{dt^2} + \xi/\mu \frac{d\Delta n_i}{dt} + \Delta n_i = n_{2i} I_{\text{pump}}(t') \tag{8}$$

where n_{2i} is the ith contribution to the nonlinear refractive index n_2. All of the noninstantaneous field-driven responses are associated with nuclear motions through the correlation function $\langle \chi_{ij}(t), \chi_{kl}(0) \rangle$ and therefore may not be capable of instantaneously following our femtosecond laser pulses. The solution to (8) in the overdamped limit is

$$\Delta n_i(t) = \frac{n_{2i}\beta_i}{(I/\mu)^{1/2}} \int_{-\infty}^{t} \sinh\left[(t - t')/\beta_i\right] \cdot \exp\left[-(t - t')/\tau_i\right] I_{\text{pump}}(t')\, dt' \tag{9}$$

where

$$\tau_i = \frac{2I}{\xi}; \qquad \beta_i = 2\left[(\xi/I)^2 - 4(\mu/I)\right]^{-1/2}$$

and naturally accounts for this behavior by including the physically required ''quadratic lag'' while maintaining the exponential diffusive character at long times. Equation (9) reduces to the diffusion result [31] in the limit of small β_i and is used to model the exponentially decaying components of the OKE response [responses 1) and 2)].

✓ Diffusive reorientational $r_3(\tau) = a_3 \exp(-\frac{\tau}{\tau_{diff}})[1 - \exp(-\frac{\tau}{\beta_3})]$

✓ Intermediate contribution $r_4(\tau) = a_4 \exp(-\frac{\tau}{\tau_{\text{int}}})[1 - \exp(-\frac{\tau}{\beta_4})]$

The solution to (8) in the underdamped limit is

$$\Delta n_i(t) = \frac{n_{2i}\beta_i}{(I/\mu)^{1/2}} \int_{-\infty}^{t} \sin\left[(t - t')/\beta_i\right] \cdot \exp\left[-(t - t')/\tau_i\right] I_{\text{pump}}(t')\, dt'. \quad (10)$$

This equation describes the oscillatory motion of a molecule in a potential well defined by the surrounding solvent structure and is used to model response 3), the ultrafast (but noninstantaneous) component of the OKE signal attributed to librational motion. The period of oscillation is given by $2\pi\beta_i$.

✓ Libration motion

$$r_2(\tau) = a_2 \exp(-\frac{\tau}{\tau_{lib}}) \exp(-\frac{\alpha^2\tau^2}{2}) \sin(\omega_0\tau)$$

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등방성 매질에서의 광학적 Kerr 효과

$$P_i^{(3)}(\omega') = \sum_{j,k,l} \chi_{ijkl}^{(3)}(\omega' = \omega' + \omega - \omega) E_j(\omega') E_k(\omega) E_l(\omega)$$

Isotropic

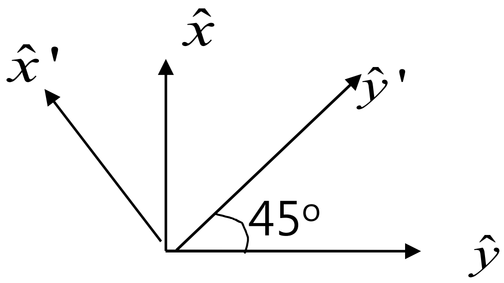
$$\chi_{1111} = \chi_{2222} = \chi_{3333}$$

$$\chi_{1122} = \chi_{1133} = \chi_{2233} = \chi_{2211} = \chi_{3311} = \chi_{3322}$$

$$\chi_{1212} = \chi_{1313} = \chi_{2323} = \chi_{2121} = \chi_{3131} = \chi_{3232}$$

$$\chi_{1221} = \chi_{1331} = \chi_{2112} = \chi_{2332} = \chi_{3113} = \chi_{3223}$$

$$\chi_{1111} = \chi_{1122} + \chi_{1212} + \chi_{1221}$$



$$\begin{bmatrix} P_x' \\ P_y' \end{bmatrix} = \begin{bmatrix} \cos 45^o & \sin 45^o \\ -\sin 45^0 & \cos 45^o \end{bmatrix} \begin{bmatrix} P_x \\ 0 \end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix} P_x \\ -P_x \end{bmatrix}$$

$$\begin{bmatrix} E_x' \\ E_y' \end{bmatrix} = \begin{bmatrix} \cos 45^o & \sin 45^o \\ -\sin 45^o & \cos 45^o \end{bmatrix} \begin{bmatrix} E_x \\ 0 \end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix} E_x \\ -E_X \end{bmatrix}$$

$$\therefore P_x' = \frac{1}{\sqrt{2}} P_x , \qquad P_y' = -\frac{1}{\sqrt{2}} P_x , \qquad E_x' = \frac{1}{\sqrt{2}} E_x , \qquad E_y = \frac{1}{\sqrt{2}} E_x$$

$$P_x' = \chi_{1111} E_x'^3 + \chi_{1122} E_x' E_y'^2 + \chi_{1221} E_x' E_y'^2 + \chi_{1212} E_x' E_y'^2$$

$$\frac{1}{\sqrt{2}} \chi_{1111}^{(3)} E_x^3 = \frac{1}{2\sqrt{2}} \chi_{1111}^{(3)} E_x^3 + \left(\chi_{1122}^{(3)} + \chi_{1221}^{(3)} + \chi_{1212}^{(3)} \right) \frac{1}{\sqrt{2}} E_x \cdot \frac{1}{2} E_x^2$$

$$2\chi_{1111}^{(3)} = \chi_{1111}^{(3)} + (\chi_{1122}^{(3)} + \chi_{1221}^{(3)} + \chi_{1212}^{(3)})$$

$$\chi_{1111}^{(3)} = \chi_{1122}^{(3)} + \chi_{1221}^{(3)} + \chi_{1212}^{(3)}$$

Pump beam $E(\omega) = E_x(\omega)$

Probe beam $E(\omega') = E_x(\omega') + E_y(\omega')$

$$P_i^{(3)}(\omega') = \sum_{j,k,l} \chi_{ijkl}^{(3)}(\omega' = \omega' + \omega - \omega) E_j(\omega') E_k(\omega) E_l(\omega)$$

$$\begin{aligned}
P_x^{(3)}(\omega') = {} & \chi_{1111}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_1(\omega) E^*{}_1(\omega) \\
& + \chi_{1112}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_1(\omega) E^*{}_2(\omega) \\
& + \chi_{1121}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_2(\omega) E^*{}_1(\omega) \\
& + \chi_{1122}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_2(\omega) E^*{}_2(\omega) \\
& + \chi_{1211}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_1(\omega) E^*{}_1(\omega) \\
& + \chi_{1212}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_1(\omega) E^*{}_2(\omega) \\
& + \chi_{1221}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_2(\omega) E^*{}_1(\omega) \\
& + \chi_{1222}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_2(\omega) E^*{}_2(\omega)
\end{aligned}$$

$$= (\chi_{1122}^{(3)} + \chi_{1212}^{(3)} + \chi_{1221}^{(3)}) E_x(\omega') E(\omega) E^*(\omega)$$

$$(\because \chi_{1111}^{(3)} = \chi_{1122}^{(3)} + \chi_{1221}^{(3)} + \chi_{1212}^{(3)})$$

$$
\begin{aligned}
P_y^{(3)}(\omega') = {} & \chi_{2111}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_1(\omega) E_1^*(\omega) \\
& + \chi_{2112}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_1(\omega) E_2^*(\omega) \\
& + \chi_{2121}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_2(\omega) E_1^*(\omega) \\
& + \chi_{2122}^{(3)}(\omega' = \omega' + \omega - \omega) E_1(\omega') E_2(\omega) E_2^*(\omega) \\
& + \chi_{2211}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_1(\omega) E_1^*(\omega) \\
& + \chi_{2212}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_1(\omega) E_2^*(\omega) \\
& + \chi_{2221}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_2(\omega) E_1^*(\omega) \\
& + \chi_{2222}^{(3)}(\omega' = \omega' + \omega - \omega) E_2(\omega') E_2(\omega) E_2^*(\omega) \\
= {} & \chi_{1122}^{(3)}(\omega' = \omega' + \omega - \omega) E_y(\omega') E(\omega) E^*(\omega)
\end{aligned}
$$

$$(\because \chi_{2211} = \chi_{1122})$$