非線形光学入門

① 吸収飽和と broadening

回転波近似

$$\tilde{\rho}_{12} = \rho_{12} e^{-i\omega t} \quad \tilde{\rho}_{21} = \rho_{21} e^{i\omega t} \quad \frac{d\tilde{\rho}_{12}}{dt} = \frac{d\rho_{12}}{dt} e^{-i\omega t} - i\omega\tilde{\rho}_{12}$$

$$\frac{d\tilde{\rho}_{12}}{dt} = i(\omega_0 - \omega)\tilde{\rho}_{12} + \frac{i}{2}\xi(\rho_{22} - \rho_{11}) - \gamma_2\tilde{\rho}_{12}$$

$$\frac{d\rho_{22}}{dt} = -\frac{i}{2}(\xi\tilde{\rho}_{21} - \xi^*\tilde{\rho}_{12}) - \gamma_1\rho_{22}$$

$$\frac{d\tilde{\rho}_{21}}{dt} = \frac{d\tilde{\rho}_{12}{}^*}{dt} \cong -i(\omega_0 - \omega)\tilde{\rho}_{21} - \frac{i}{2}\xi^*(\rho_{22} - \rho_{11}) - \gamma_2\tilde{\rho}_{21}$$

$$\frac{d\rho_{11}}{dt} = -\frac{d\rho_{22}}{dt} \cong \frac{i}{2}(\xi\tilde{\rho}_{21} - \xi^*\tilde{\rho}_{12}) + \gamma_1\rho_{22}$$

定常解 $\dfrac{d\tilde{\rho}_{12}}{dt} = 0 \quad \dfrac{d\rho_{22}}{dt} = 0$

$$\frac{d\tilde{\rho}_{12}}{dt} = [i(\omega_0 - \omega) - \gamma_2]\tilde{\rho}_{12} + \frac{i}{2}\xi(\rho_{22} - \rho_{11}) = 0$$

$$\frac{d\rho_{22}}{dt} = -\frac{i}{2}(\xi\tilde{\rho}_{21} - \xi^*\tilde{\rho}_{12}) - \gamma_1\rho_{22} = 0$$

$$\tilde{\rho}_{12} = \frac{-\frac{i}{2}\xi(\rho_{22} - \rho_{11})}{i(\omega_0 - \omega) - \gamma_2} = \frac{-\frac{i}{2}\xi(2\rho_{22} - 1)}{i(\omega_0 - \omega) - \gamma_2}$$

$$\gamma_1\rho_{22} = -\frac{i}{2}(\xi\tilde{\rho}_{21} - \xi^*\tilde{\rho}_{12}) = -\frac{i}{2}\left[\xi\frac{\frac{i}{2}\xi^*(2\rho_{22} - 1)}{-i(\omega_0 - \omega) - \gamma_2} - \xi^*\frac{-\frac{i}{2}\xi(2\rho_{22} - 1)}{i(\omega_0 - \omega) - \gamma_2}\right]$$

$$= \frac{|\xi|^2}{4}(2\rho_{22} - 1)\left[\frac{1}{-i(\omega_0 - \omega) - \gamma_2} + \frac{1}{i(\omega_0 - \omega) - \gamma_2}\right]$$

$$= \frac{|\xi|^2}{4}(2\rho_{22} - 1)\frac{[i(\omega_0 - \omega) - \gamma_2] + [-i(\omega_0 - \omega) - \gamma_2]}{(\omega_0 - \omega)^2 + \gamma_2{}^2}$$

$$= \frac{|\xi|^2}{4}(2\rho_{22} - 1)\frac{-2\gamma_2}{(\omega_0 - \omega)^2 + \gamma_2{}^2}$$

$$\gamma_1\rho_{22} = -|\xi|^2\frac{\gamma_2}{(\omega_0 - \omega)^2 + \gamma_2{}^2}\rho_{22} + \frac{|\xi|^2}{2}\frac{\gamma_2}{(\omega_0 - \omega)^2 + \gamma_2{}^2}$$

$$\gamma_1[(\omega_0-\omega)^2+\gamma_2{}^2]\rho_{22}=-|\xi|^2\gamma_2\rho_{22}+\frac{|\xi|^2}{2}\gamma_2$$

$$\left\{\gamma_1[(\omega_0-\omega)^2+\gamma_2{}^2]+|\xi|^2\gamma_2\right\}\rho_{22}=\frac{|\xi|^2}{2}\gamma_2$$

$$\rho_{22}=\frac{\frac{|\xi|^2}{2}\frac{\gamma_2}{\gamma_1}}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}\to\frac{1}{2}\quad(|\xi|\to\infty)$$ 最大でも$\frac{1}{2}$、つまり二準位系では反転分布を作れない

$$\tilde{\rho}_{12}=\frac{-\frac{i}{2}\xi}{i(\omega_0-\omega)-\gamma_2}(2\rho_{22}-1)$$

$$=\frac{-\frac{i}{2}\xi}{i(\omega_0-\omega)-\gamma_2}\frac{|\xi|^2\frac{\gamma_2}{\gamma_1}-(\omega_0-\omega)^2-\gamma_2{}^2-|\xi|^2\frac{\gamma_2}{\gamma_1}}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}$$

$$=\frac{\frac{i}{2}\xi}{i(\omega_0-\omega)-\gamma_2}\frac{(\omega_0-\omega)^2+\gamma_2{}^2}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}$$

$$=\frac{i}{2}\xi\frac{-i(\omega_0-\omega)-\gamma_2}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}$$

$$\rho_{12}=\tilde{\rho}_{12}e^{i\omega t}=\frac{i}{2}\frac{\mu_{12}E_0}{\hbar}\frac{-i(\omega_0-\omega)-\gamma_2}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}e^{i\omega t}$$

巨視的分極

$$P(t)=\frac{N}{V}(\rho_{21}\mu_{12}+\rho_{12}\mu_{21})$$

$$=\frac{N}{V}\left[\left(-\frac{i}{2}\frac{\mu_{21}E_0}{\hbar}\frac{i(\omega_0-\omega)-\gamma_2}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}e^{-i\omega t}\right)\mu_{12}+\left(\frac{i}{2}\frac{\mu_{12}E_0}{\hbar}\frac{-i(\omega_0-\omega)-\gamma_2}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}e^{i\omega t}\right)\mu_{21}\right]$$

$$=\varepsilon_0\frac{E_0}{2}\left[\frac{N}{V}\frac{|\mu_{12}|^2}{\varepsilon_0\hbar}\left(\frac{(\omega_0-\omega)+i\gamma_2}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}\right)e^{-i\omega t}+\frac{N}{V}\frac{|\mu_{12}|^2}{\varepsilon_0\hbar}\left(\frac{(\omega_0-\omega)-i\gamma_2}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}\right)e^{i\omega t}\right]$$

$$=\varepsilon_0\frac{E_0}{2}\left[\chi(\omega)e^{-i\omega t}+\chi(-\omega)e^{i\omega t}\right]\qquad E(t)=\frac{E_0}{2}(e^{-i\omega t}+e^{i\omega t})$$

$$\chi(\omega)=\frac{N}{V}\frac{|\mu_{12}|^2}{\varepsilon_0\hbar}\frac{(\omega_0-\omega)+i\gamma_2}{(\omega_0-\omega)^2+\gamma_2{}^2+|\xi|^2\frac{\gamma_2}{\gamma_1}}$$ 光電場の増大による吸収飽和、broadeningを表す非線形感受率

[補足：$e^{i\omega t}$の係数$\chi(-\omega)$が上の$\chi(\omega)$で$\omega\to-\omega$としたものになっていないのは回転波近似を使っているから
＝ρ_{12}に$e^{i\omega t}$と$e^{-i\omega t}$の２つの項があるはずが、回転波近似で後者を落としている]

$$\chi(\omega)=\frac{N}{V}\frac{|\mu_{12}|^2}{\varepsilon_0\hbar}\frac{(\omega_0-\omega)+i\gamma_2}{(\omega_0-\omega)^2+{\gamma_2}^2+|\frac{\mu_{12}E_0}{\hbar}|^2\frac{\gamma_2}{\gamma_1}}=\frac{N}{V}\frac{|\mu_{12}|^2}{\varepsilon_0\hbar}\frac{(\omega_0-\omega)+i\gamma_2}{[(\omega_0-\omega)^2+{\gamma_2}^2]\left[1+\frac{|\frac{\mu_{12}E_0}{\hbar}|^2\frac{\gamma_2}{\gamma_1}}{(\omega_0-\omega)^2+{\gamma_2}^2}\right]}$$

$$\approx\frac{N}{V}\frac{|\mu_{12}|^2}{\varepsilon_0\hbar}\frac{(\omega_0-\omega)+i\gamma_2}{[(\omega_0-\omega)^2+{\gamma_2}^2]}\left[1-\frac{|\frac{\mu_{12}E_0}{\hbar}|^2\frac{\gamma_2}{\gamma_1}}{(\omega_0-\omega)^2+{\gamma_2}^2}\right]=\frac{N}{V}\frac{|\mu_{12}|^2}{\varepsilon_0\hbar}\frac{1}{(\omega_0-\omega)-i\gamma_2}\left[1-\frac{|\frac{\mu_{12}E_0}{\hbar}|^2\frac{\gamma_2}{\gamma_1}}{(\omega_0-\omega)^2+{\gamma_2}^2}\right]$$

$$=\frac{N}{V}\frac{|\mu_{12}|^2}{\varepsilon_0\hbar}\frac{1}{(\omega_0-\omega)-i\gamma_2}-\frac{N}{V}\frac{|\mu_{12}|^2}{\varepsilon_0\hbar}|\frac{\mu_{12}E_0}{\hbar}|^2\frac{1}{(\omega_0-\omega)-i\gamma_2}\frac{\frac{\gamma_2}{\gamma_1}}{(\omega_0-\omega)^2+{\gamma_2}^2}$$

$$=\frac{1}{\varepsilon_0}\frac{N}{V}\frac{|\mu_{12}|^2}{\hbar}\frac{1}{(\omega_0-\omega)-i\gamma_2}-\frac{1}{\varepsilon_0}\frac{N}{V}\frac{|\mu_{12}|^4}{\hbar^3}\frac{1}{(\omega_0-\omega)-i\gamma_2}\frac{\frac{\gamma_2}{\gamma_1}}{(\omega_0-\omega)^2+{\gamma_2}^2}|E_0|^2$$

$$E(t)=\frac{E_0}{2}(e^{-i\omega t}+e^{i\omega t})$$

$$E^3=\frac{E_0}{2}(e^{-i\omega t}+e^{i\omega t})\frac{E_0}{2}(e^{-i\omega t}+e^{i\omega t})\frac{E_0}{2}(e^{-i\omega t}+e^{i\omega t})$$

$$=(\frac{E_0}{2})^3(e^{-i3\omega t}+3e^{-i\omega t}+3e^{i\omega t}+e^{i3\omega t})$$

上の扱いでは回転波近似により$3\omega,-3\omega$を発生する項を無視している

$P(t)=\varepsilon_0\frac{E_0}{2}\left[\chi(\omega)e^{-i\omega t}+\chi(-\omega)e^{i\omega t}\right]=\varepsilon_0\frac{E_0}{2}\left[\chi(\omega;-\omega)e^{-i\omega t}+\chi(-\omega;\omega)e^{i\omega t}\right]$と同様に

$$P_{-\omega}{}^{(3)}(t)=\varepsilon_0(\frac{E_0}{2})^3[\chi^{(3)}(\omega;-\omega,\omega,-\omega)e^{-i\omega t}+\chi^{(3)}(\omega;-\omega,-\omega,\omega)e^{-i\omega t}+\chi^{(3)}(\omega;\omega,-\omega,-\omega)e^{-i\omega t}]$$

$P_{\omega}{}^{(3)}(t)=\varepsilon_0(\frac{E_0}{2})^3[\chi^{(3)}(-\omega;\omega,-\omega,\omega)e^{i\omega t}+\chi^{(3)}(-\omega;\omega,\omega,-\omega)e^{i\omega t}+\chi^{(3)}(-\omega;-\omega,\omega,\omega)e^{i\omega t}]$で$\chi^{(3)}(\omega)$を定義すると、

$$\chi^{(3)}(\omega;-\omega,\omega,-\omega)+\chi^{(3)}(\omega;-\omega,-\omega,\omega)+\chi^{(3)}(\omega;\omega,-\omega,-\omega)=-4\frac{1}{\varepsilon_0}\frac{N}{V}\frac{|\mu_{12}|^4}{\hbar^3}\frac{1}{(\omega_0-\omega)-i\gamma_2}\frac{\frac{\gamma_2}{\gamma_1}}{(\omega_0-\omega)^2+{\gamma_2}^2}$$

$\omega=+\omega-\omega+\omega$

$\chi^{(3)}(-\omega;\omega,-\omega,\omega)$:　$-\omega+\omega-\omega+\omega=0$　と書く場合が多い　$\chi^{(1)}(-\omega;\omega)$

② 二倍波発生

二倍波発生（第2高調波発生）、光整流

$$E(t) = E_0 \cos \omega t = \frac{E_0}{2}(e^{-i\omega t} + e^{i\omega t})$$

$$P^{(2)} = \varepsilon_0 \chi^{(2)} E^2 = \varepsilon_0 \chi^{(2)} {E_0}^2 \cos^2 \omega t = \varepsilon_0 \chi^{(2)} {E_0}^2 \frac{1+\cos 2\omega t}{2}$$ 第１項：光整流、第２項：二倍波発生

$$P^{(2)} = \varepsilon_0 \chi^{(2)} E^2 = \varepsilon_0 \chi^{(2)} \frac{{E_0}^2}{4}(e^{-i\omega t} + e^{i\omega t})(e^{-i\omega t} + e^{i\omega t}) = \varepsilon_0 \chi^{(2)} \frac{{E_0}^2}{4}(2 + e^{-i2\omega t} + e^{i2\omega t})$$

和周波発生、差周波発生

$$E_1(t) = E_{10} \cos \omega_1 t = \frac{E_{10}}{2}(e^{-i\omega_1 t} + e^{i\omega_1 t}), E_2(t) = E_{20} \cos \omega_2 t = \frac{E_{20}}{2}(e^{-i\omega_2 t} + e^{i\omega_2 t})$$

$$P^{(2)} = \varepsilon_0 \chi^{(2)} E_1 E_2 = \varepsilon_0 \chi^{(2)} E_{10} E_{20} \cos \omega_1 t \cos \omega_2 t = \varepsilon_0 \chi^{(2)} E_{10} E_{20} \frac{1}{2}[\cos(\omega_1 + \omega_2)t + \cos(\omega_1 - \omega_2)t]$$

$$P^{(2)} = \varepsilon_0 \chi^{(2)} E_1 E_2 = \varepsilon_0 \chi^{(2)} \frac{E_{10}}{2}\frac{E_{20}}{2}(e^{-i\omega_1 t} + e^{i\omega_1 t})(e^{-i\omega_2 t} + e^{i\omega_2 t})$$

$$= \varepsilon_0 \chi^{(2)} \frac{E_{10}}{2}\frac{E_{20}}{2}(e^{-i(\omega_1+\omega_2)t} + e^{i(\omega_1+\omega_2)t} + e^{-i(\omega_1-\omega_2)t} + e^{i(\omega_1-\omega_2)t})$$

実際には、物質中での非線形分極発生過程をきちんと考えると

$$E_1(t) = E_{10} e^{-i\omega_1 t} \quad E_2(t) = E_{20} e^{-i\omega_2 t}$$

$$P^{(2)} = \varepsilon_0 [\chi^{(2)}(\omega_1 + \omega_2) E_1 E_2 + \chi^{(2)}(-\omega_1 - \omega_2) {E_1}^* {E_2}^* + \chi^{(2)}(\omega_1 - \omega_2) E_1 {E_2}^* + \chi^{(2)}(-\omega_1 + \omega_2) {E_1}^* E_2]$$

$$P_i = {\chi_{ijk}}^{(2)} E_j E_k$$

座標を反転

このとき、ベクトルの符号は反転するが、系に反転対称性があれば${\chi_{ijk}}^{(2)}$は不変

$$-P_i = {\chi_{ijk}}^{(2)}(-E_j)(-E_k) = {\chi_{ijk}}^{(2)} E_j E_k$$

したがって$0 = {\chi_{ijk}}^{(2)} E_j E_k$

任意のE_j, E_kで成り立つので${\chi_{ijk}}^{(2)} = 0$

2次の非線形感受率がゼロでない物質の必要条件　反転対称性がないこと

2次の非線形光学結晶　$\mathrm{LiIO_3, KTP(KTiOPO_4), KDP(KH_2PO_4), LBO(LiB_3O_5), BBO(\beta\text{-}BaB_2O_4)}$　$\omega, 2\omega$で透明

$\mathrm{GaAs, ZnS}$

$$\begin{cases} \nabla \times \boldsymbol{B} = \mu_0 \dfrac{\partial}{\partial t}(\varepsilon_0 \boldsymbol{E} + \boldsymbol{P}) \\ \nabla \times \boldsymbol{E} = -\dfrac{\partial \boldsymbol{B}}{\partial t} \end{cases}$$

$$\nabla \times (\nabla \times \boldsymbol{E}) = \nabla \times (-\frac{\partial \boldsymbol{B}}{\partial t}) = -\frac{\partial}{\partial t}(\nabla \times \boldsymbol{B}) = -\mu_0 \frac{\partial^2}{\partial t^2}(\varepsilon_0 \boldsymbol{E} + \boldsymbol{P}) = -\mu_0 \frac{\partial^2}{\partial t^2}(\varepsilon \boldsymbol{E} + \boldsymbol{P}_{\mathrm{NL}})$$

$$\nabla \times (\nabla \times \boldsymbol{E}) = -\nabla^2 \boldsymbol{E} \quad \because \nabla \cdot \boldsymbol{E} = 0$$

$$\boldsymbol{P} = \varepsilon_0 \chi_{\mathrm{L}} \boldsymbol{E} + \boldsymbol{P}_{\mathrm{NL}}$$

$$\varepsilon = \varepsilon_0 (1 + \chi_{\mathrm{L}})$$

$$\nabla^2 \boldsymbol{E} = \mu_0 \frac{\partial^2}{\partial t^2}(\varepsilon \boldsymbol{E} + \boldsymbol{P}_{\mathrm{NL}})$$

$$\begin{cases} \nabla\times\boldsymbol{B} = \mu_0 \dfrac{\partial}{\partial t}(\varepsilon_0\boldsymbol{E}+\boldsymbol{P}) \\ \nabla\times\boldsymbol{E} = -\dfrac{\partial \boldsymbol{B}}{\partial t} \end{cases}$$

$$\nabla\times(\nabla\times\boldsymbol{E}) = \nabla\times(-\frac{\partial\boldsymbol{B}}{\partial t}) = -\frac{\partial}{\partial t}(\nabla\times\boldsymbol{B}) = -\mu_0\frac{\partial^2}{\partial t^2}(\varepsilon_0\boldsymbol{E}+\boldsymbol{P}) = -\mu_0\frac{\partial^2}{\partial t^2}(\varepsilon\boldsymbol{E}+\boldsymbol{P}_{\mathrm{NL}})$$

$$\nabla\times(\nabla\times\boldsymbol{E}) = -\nabla^2\boldsymbol{E} \quad \because \nabla\cdot\boldsymbol{E}=0$$

$$\boldsymbol{P} = \varepsilon_0\chi_{\mathrm{L}}\boldsymbol{E}+\boldsymbol{P}_{\mathrm{NL}}$$

$$\varepsilon = \varepsilon_0(1+\chi_{\mathrm{L}})$$

入射電磁波（基本波）$\boldsymbol{E}_\omega(t) = \boldsymbol{E}_{\omega 0}\cos(\omega t - k_{(\omega)}z) = \dfrac{\boldsymbol{E}_{\omega 0}}{2}(e^{-i(\omega t-kz)}+e^{i(\omega t-kz)})$

による2次の非線形分極 $\boldsymbol{P}_{\mathrm{NL}} = \boldsymbol{P}^{(2)} = \varepsilon_0\chi^{(2)}\dfrac{\boldsymbol{E}_0{}^2}{2}\cos(2\omega t-2k_{(\omega)}z)$

$$\nabla^2\boldsymbol{E} = \mu_0\frac{\partial^2}{\partial t^2}(\varepsilon\boldsymbol{E}+\boldsymbol{P}_{\mathrm{NL}})$$

発生二倍波 $\boldsymbol{E} = \boldsymbol{E}_0(z)\cos(2\omega t - k_{(2\omega)}z)$

$$\frac{\partial^2}{\partial z^2}\boldsymbol{E} - \mu_0\varepsilon\frac{\partial^2}{\partial t^2}\boldsymbol{E} = \mu_0\frac{\partial^2}{\partial t^2}\boldsymbol{P}_{\mathrm{NL}} \quad \mathrm{(A)-(B)=(C)}$$

$$\mathrm{(A)}\ \frac{\partial^2}{\partial z^2}\boldsymbol{E} = \frac{\partial^2}{\partial z^2}[\boldsymbol{E}_0(z)\cos(2\omega t-k_{(2\omega)}z)] = \frac{\partial^2}{\partial z^2}[\frac{\boldsymbol{E}_0(z)}{2}(e^{-i(2\omega t-k_{(2\omega)}z)}+e^{i(2\omega t-k_{(2\omega)}z)})]$$

$$= \frac{1}{2}\frac{d^2\boldsymbol{E}_0(z)}{dz^2}e^{-i(2\omega t-k_{(2\omega)}z)} + \frac{d\boldsymbol{E}_0(z)}{dz}(ik_{(2\omega)})e^{-i(2\omega t-k_{(2\omega)}z)} + \frac{\boldsymbol{E}_0(z)}{2}(-k_{(2\omega)}{}^2)e^{-i(2\omega t-k_{(2\omega)}z)} + \mathrm{C.C.}$$

$$\frac{d\boldsymbol{E}_0}{dz}k_{(2\omega)} >> \frac{d^2\boldsymbol{E}_0}{dz^2} \quad \text{slowly varying envelope approx.}$$

$$\cong -\frac{1}{2}\left[k_{(2\omega)}{}^2\boldsymbol{E}_0(z) - 2ik_{(2\omega)}\frac{d\boldsymbol{E}_0(z)}{dz}\right]e^{-i(2\omega t-k_{(2\omega)}z)} + \mathrm{C.C.}$$

$$\mathrm{(B)}\ \mu_0\varepsilon\frac{\partial^2}{\partial t^2}\boldsymbol{E} = -\mu_0\varepsilon(2\omega)^2\frac{\boldsymbol{E}_0(z)}{2}e^{-i(2\omega t-k_{(2\omega)}z)} + \mathrm{C.C.}$$

$$\mathrm{(C)}\ \mu_0\frac{\partial^2}{\partial t^2}\boldsymbol{P}_{\mathrm{NL}} = -\mu_0\varepsilon_0(2\omega)^2\chi^{(2)}\frac{\boldsymbol{E}_{\omega 0}{}^2}{4}e^{-i(2\omega t-2k_{(\omega)}z)} + \mathrm{C.C.}$$

$\mu_0\varepsilon(2\omega)^2 = k_{(2\omega)}{}^2$の関係式を使って

$$ik_{(2\omega)}\frac{d\boldsymbol{E}_0(z)}{dz}e^{-i(2\omega t-k_{(2\omega)}z)} + \mathrm{C.C.} = -\mu_0\varepsilon_0(2\omega)^2\chi^{(2)}\frac{\boldsymbol{E}_{\omega 0}{}^2}{4}e^{-i(2\omega t-2k_{(\omega)}z)} + \mathrm{C.C.}$$

$k_{(2\omega)} = 2\omega\sqrt{\mu_0\varepsilon}$より

$$\frac{d\boldsymbol{E}_0(z)}{dz} = i\sqrt{\mu_0}\frac{\varepsilon_0}{\sqrt{\varepsilon}}(2\omega)\chi^{(2)}\frac{\boldsymbol{E}_{\omega 0}{}^2}{4}e^{i(2k_{(\omega)}-k_{(2\omega)})z} \equiv iAe^{i(2k_{(\omega)}-k_{(2\omega)})z}$$

no 2ω input

$E_0(0)=0 \quad 0-L$

$$E_0(L) = A\frac{e^{i(2k_{(\omega)}-k_{(2\omega)})L}-1}{2k_{(\omega)}-k_{(2\omega)}} = Ae^{i\frac{2k_{(\omega)}-k_{(2\omega)}}{2}L}\frac{e^{i\frac{2k_{(\omega)}-k_{(2\omega)}}{2}L}-e^{-i\frac{2k_{(\omega)}-k_{(2\omega)}}{2}L}}{2k_{(\omega)}-k_{(2\omega)}} = 2iAe^{i\frac{2k_{(\omega)}-k_{(2\omega)}}{2}L}\frac{\sin(\frac{2k_{(\omega)}-k_{(2\omega)}}{2}L)}{2k_{(\omega)}-k_{(2\omega)}}$$

$$|E_0(L)|^2 = A^2\frac{\sin^2(\frac{2k_{(\omega)}-k_{(2\omega)}}{2}L)}{(\frac{2k_{(\omega)}-k_{(2\omega)}}{2})^2}$$

位相整合(phase matching)条件　$2k_{(\omega)} - k_{(2\omega)} = 0$　　$\boldsymbol{k}_{(\omega)} + \boldsymbol{k}_{(\omega)} = \boldsymbol{k}_{(2\omega)}$　$\boldsymbol{k}_{(\omega_1)} + \boldsymbol{k}_{(\omega_2)} = \boldsymbol{k}_{(\omega_1+\omega_2)}$　（運動量保存則）

を満たすとき、 $|E_0(L)|^2 = A^2 L^2$　信号最大

$$2k_{(\omega)} = k_{(2\omega)}$$

$$\frac{2\omega}{2k_{(\omega)}} = \frac{2\omega}{k_{(2\omega)}}$$

位相速度 $v = \dfrac{\omega}{k}$ より

$$v_{(\omega)} = v_{(2\omega)}$$

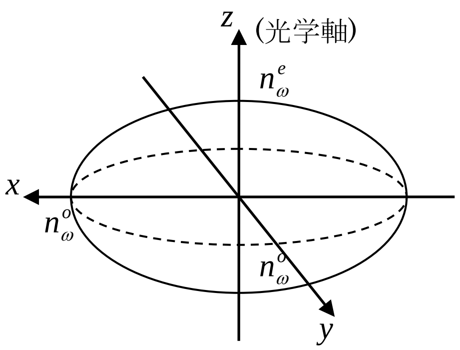
$$\frac{c}{n_{(\omega)}} = \frac{c}{n_{(2\omega)}}$$

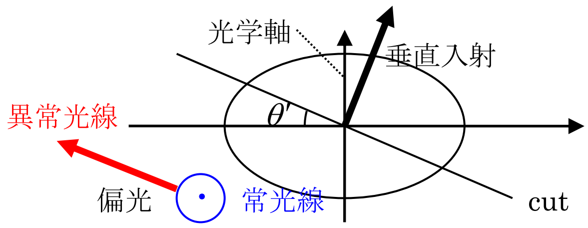
$$n_{(\omega)} = n_{(2\omega)}$$

しかし、通常　$n_{(\omega)} < n_{(2\omega)}$

どのようにすれば位相整合条件を満たせるか？

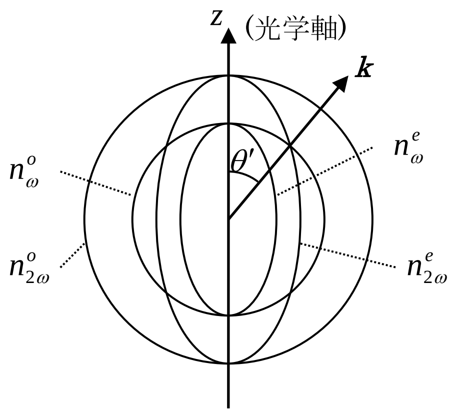
一軸性結晶(屈折率の異方性をもつ)を利用





屈折率楕円体　$\dfrac{x^2}{n^{o\,2}_\omega} + \dfrac{y^2}{n^{o\,2}_\omega} + \dfrac{z^2}{n^{e\,2}_\omega} = 1$、光学軸に垂直($xy$ 面内)に偏光している光は屈折率 n^o_ω で伝播(ordinary ray　常光線)、

光学軸と進行方向を含む平面内で偏光する光 $E_0(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ は屈折率 $n = \sqrt{n^{o\,2}_\omega \sin^2\theta + n^{e\,2}_\omega \cos^2\theta}$ で伝播 (extra ordinary ray　異常光線)



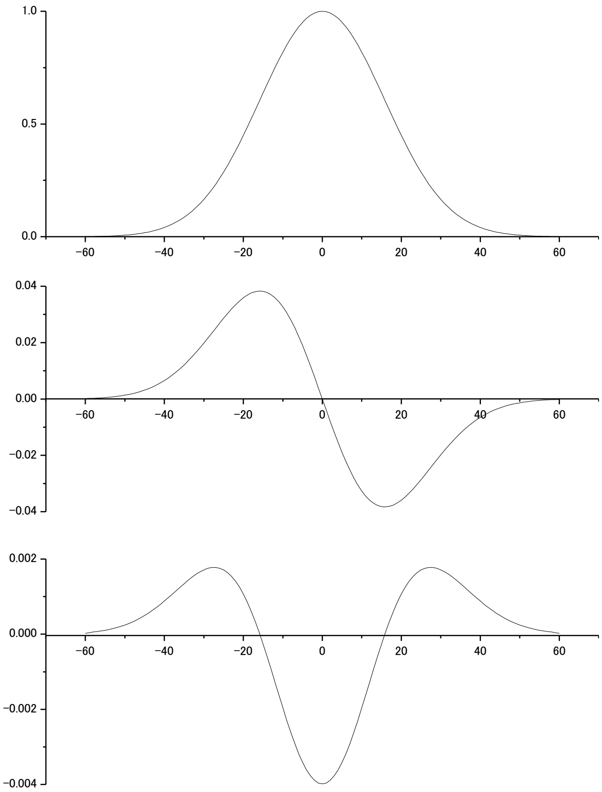
③ 光整流

$\chi^{(2)}(0,-\omega,\omega)$　**フェムト秒パルスによるTHz波発生のメカニズムの一つ**

$$E \propto \frac{d^2P(t)}{dt^2} \propto \frac{dI(t)}{dt}$$

フェムト秒パルス　$E(t) = E_0(t)e^{-i\omega t}$

$$P^{(2)}(t) = \varepsilon_0 \chi^{(2)}(0,-\omega,\omega)E^*E \propto |E_0(t)|^2$$

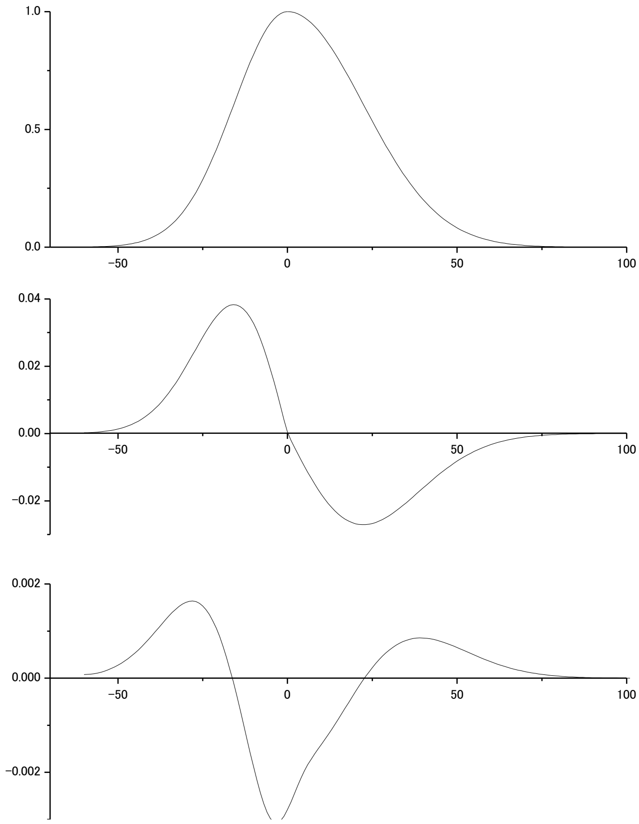


$P(t)$

$\frac{dP}{dt}$

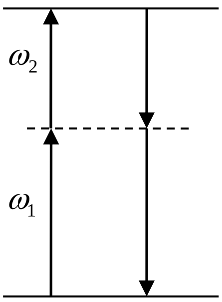
$\frac{d^2P}{dt^2}$

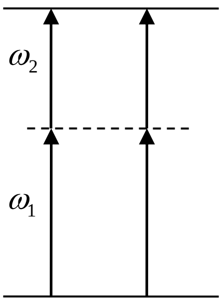
分極が瞬時応答でないとき



④ parametric down conversion　$2\omega \rightarrow \omega + \omega$

⑤ 二光子吸収　　励起子分子の生成　$\chi^{(3)}(\omega_1,-\omega_1,\omega_2,-\omega_2)$





⑥ 自己位相変調　self-phase modulation

$n = n_0 + n_1 E + n_2 EE^*$　**第2項　2次の非線形光学効果**（Pockels**効果**）**反転対称性があるとき**0

第3項　3次の非線形光学効果（kerr**効果**）self focusing **自己収束　　強度が強いほど屈折率大**

$$k = n\frac{\omega_0}{c}$$

$$E(t) = E_0(t)e^{-i(\omega_0 t - kx)} = E_0(t)e^{-i(\omega_0 t - n\frac{\omega_0}{c}x)}$$

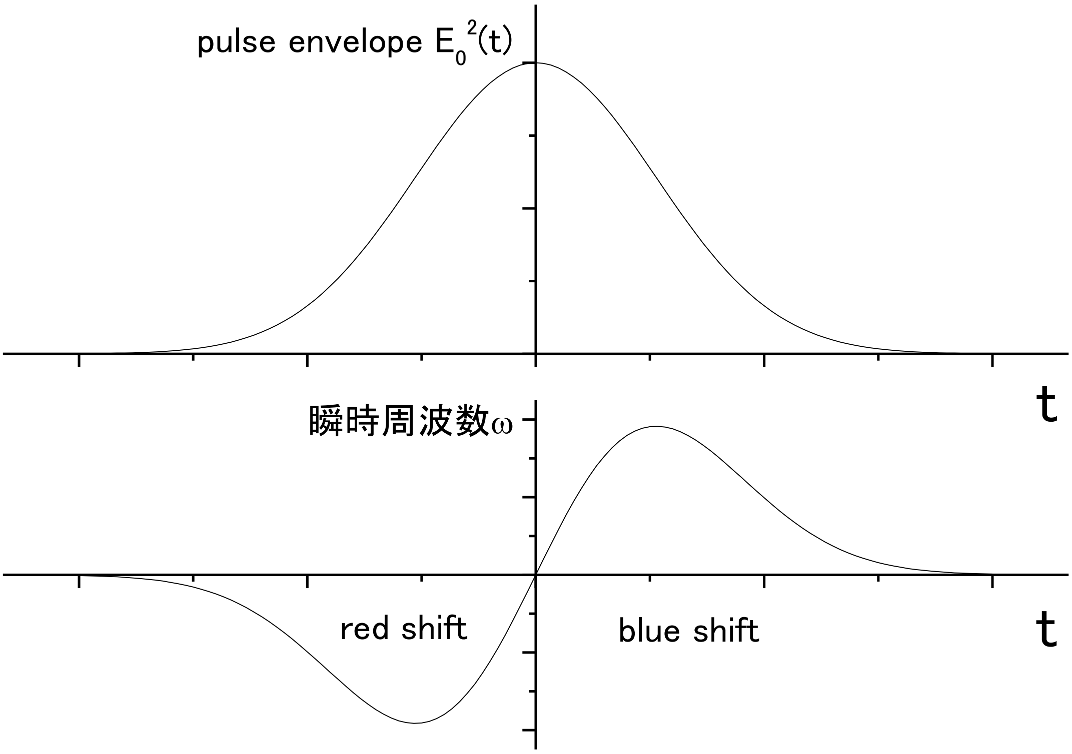
$$= E_0(t)e^{-i\omega_0 t + in_0\frac{\omega_0}{c}x + in_2|E|^2\frac{\omega_0}{c}x}$$

$$= E_0(t)e^{-i\phi(t)}$$

$$\phi(t) = \omega_0 t - n_0\frac{\omega_0}{c}x - n_2|E(t)|^2\frac{\omega_0}{c}x$$

瞬時周波数

$$\omega = \frac{d\phi(t)}{dt} = \omega_0 - n_2\frac{\omega_0}{c}x\frac{dE_0{}^2(t)}{dt}$$

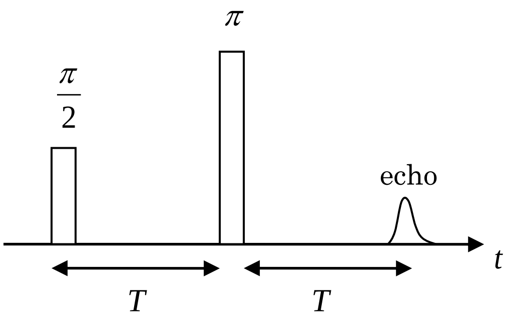


超短パルス(femto 秒～pico 秒)で行うと白色光発生（continuum generation）

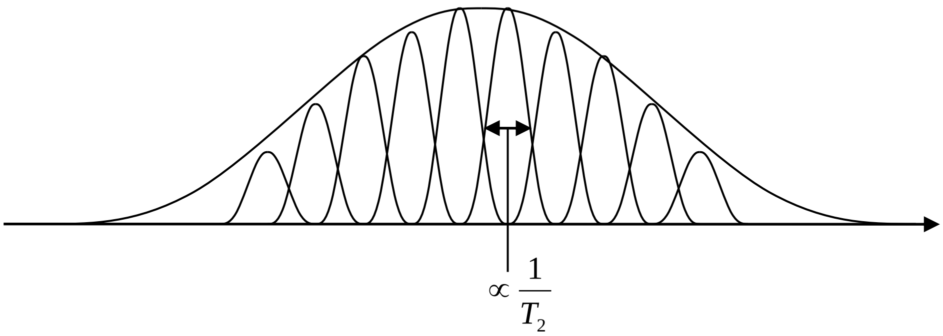
100fs, 1μJ 程度のパルスを tight に集光すれば 1mm 程度の透明媒質（ガラス、水、有機溶媒）で白色光発生

⑦ Photon Echo フォトンエコー　⊂　四光波混合　Four-Wave Mixing（３次非線形効果）

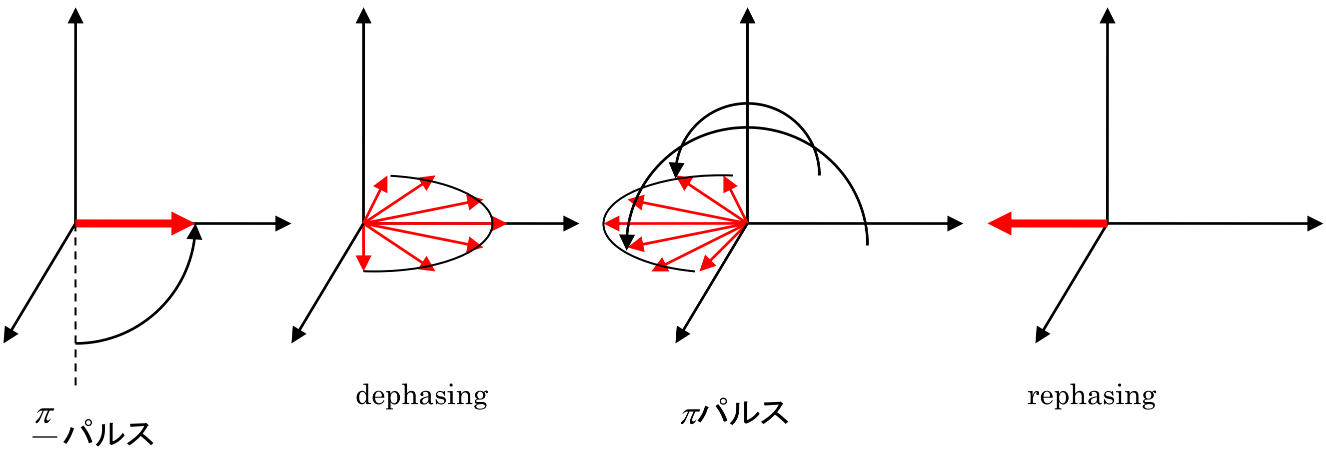
パルス面積　$\Theta = \frac{\mu_{12}}{\hbar}\int_{-\infty}^{\infty} E_0(t)dt = \int_{-\infty}^{\infty} \xi(t)dt$



エコーを観測するには吸収が不均一広がりを持つことが条件
例：熱運動している気体　ドップラー広がり
　固体中の局在中心　　個々の環境の違い
⇔　均一広がり、均一幅
　　衝突広がり、自然幅

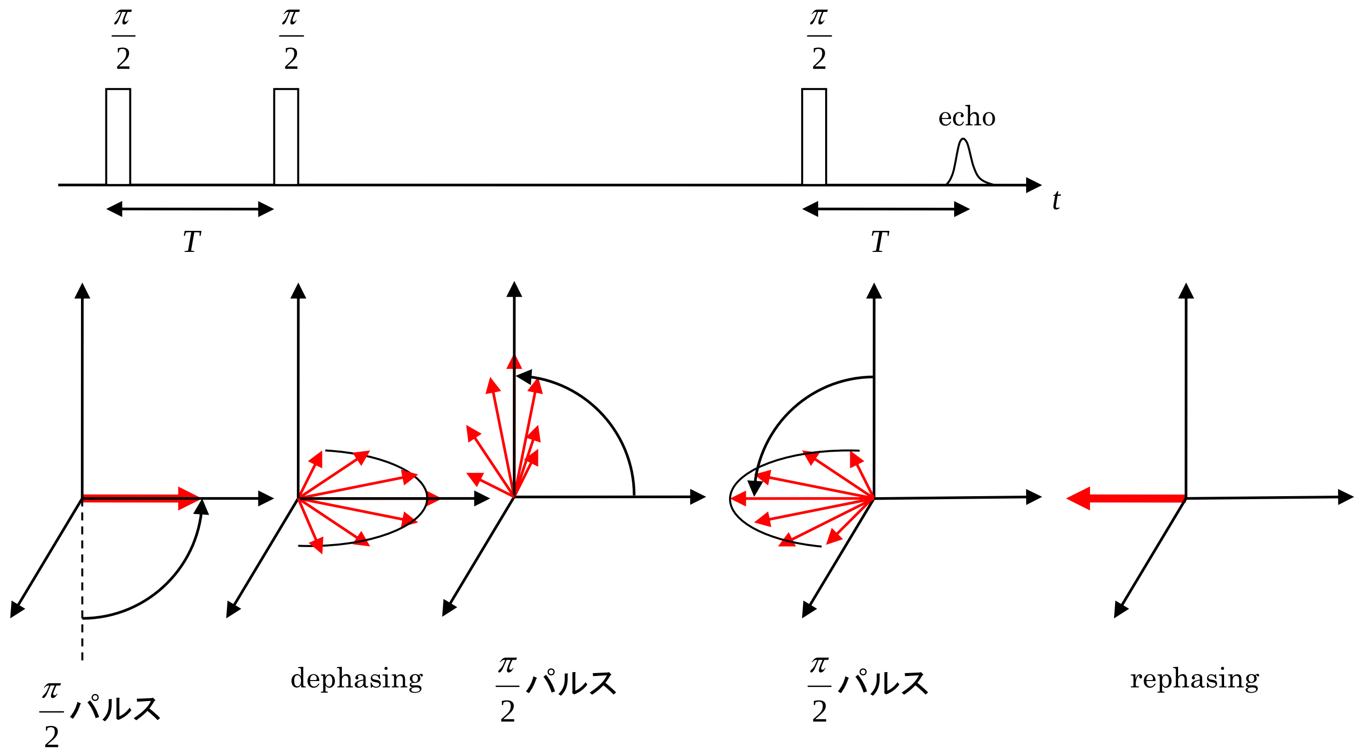


吸収線幅からは T_2（均一幅の逆数、位相緩和時間）はわからない



$\frac{\pi}{2}$ パルス

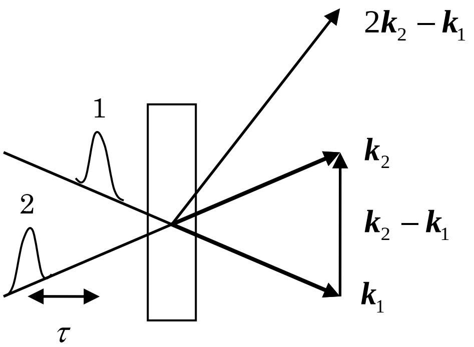
Stimulated photon echo



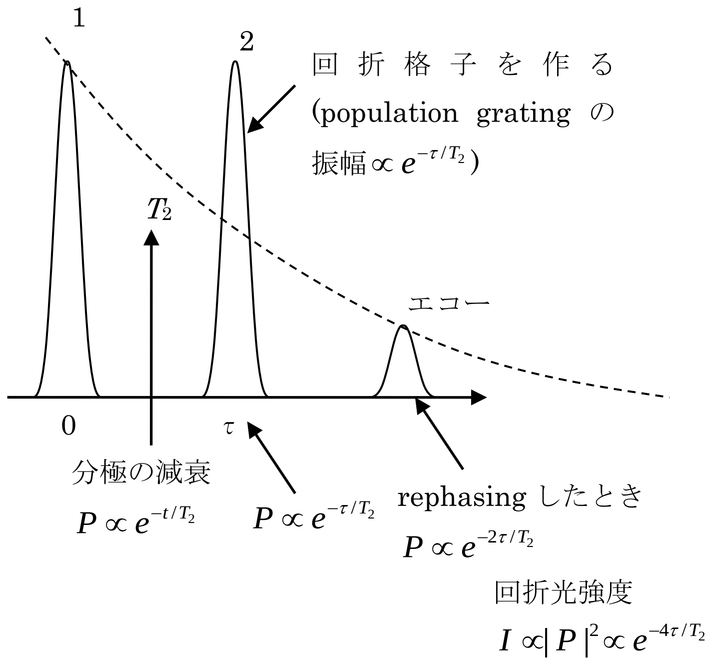
$T_1 >> T_2$

励起パルスが π/2 パルスでなくてもよい。ただし、信号の振幅が $\sin\Theta_1 \cdot \sin\Theta_2 \cdot \sin\Theta_3$ だけ小さくなる

Transient grating 法

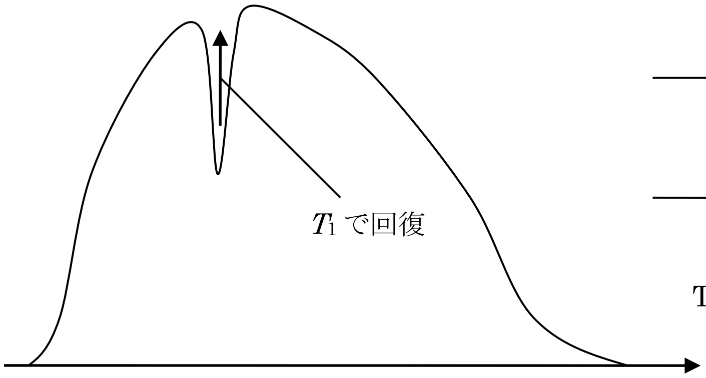


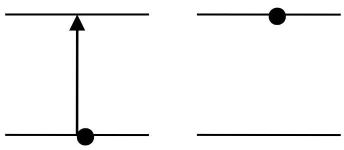
第 1,第 2 パルスの遅延時間 τ の関数として回折光強度をプロット



不均一幅から均一幅を測定する周波数領域の非線形分光

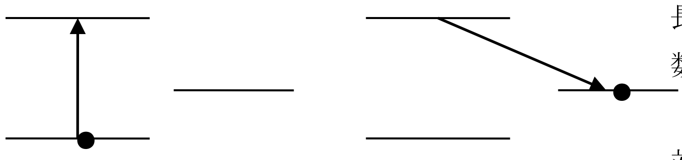
Spectral hole burning　均一幅より狭帯域の（線幅の狭い）レーザー光を使う





吸収飽和

Transient hole burning



長寿命の中間状態(特に低温で)

数時間も続くことも(永続的 hole burning)

persistent

対応する時間領域の方法

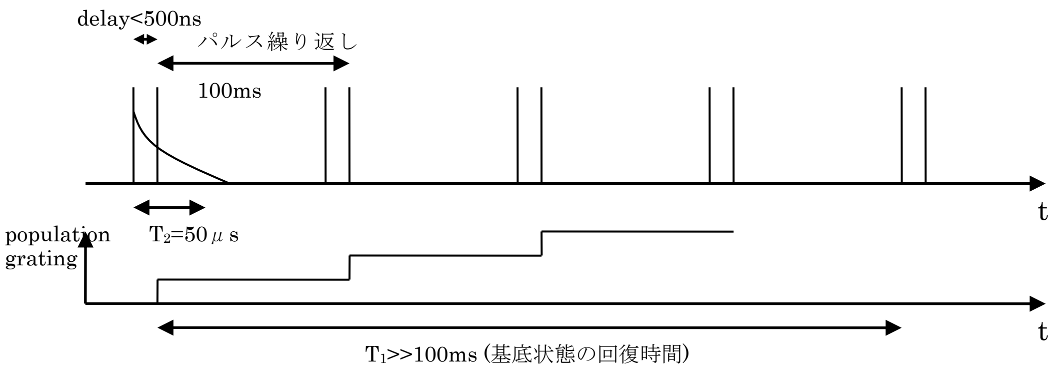
accumulated photon echo

発展：

波長多重メモリー

時間領域ホログラフィー

蓄積フォトンエコー



空間領域ホログラフィー

物体光　$E_O = E_1\cos(\boldsymbol{k}_1\cdot\boldsymbol{x}-\omega t+\phi)$

参照光　$E_R = E_2\cos(\boldsymbol{k}_2\cdot\boldsymbol{x}-\omega t)$

$$(E_O+E_R)^2 = E_1^{\ 2}\frac{1+\cos 2(\boldsymbol{k}_1\cdot\boldsymbol{x}-\omega t+\phi)}{2}+E_2^{\ 2}\frac{1+\cos 2(\boldsymbol{k}_2\cdot\boldsymbol{x}-\omega t)}{2}$$

$$+E_1E_2\{\cos[(\boldsymbol{k}_1+\boldsymbol{k}_2)\cdot\boldsymbol{x}-2\omega t+\phi)]+\cos[(\boldsymbol{k}_1-\boldsymbol{k}_2)\cdot\boldsymbol{x}+\phi)]\}$$

$$(\text{サイクル平均}) = \frac{E_1^{\ 2}}{2}+\frac{E_2^{\ 2}}{2}+E_1E_2\cos[(\boldsymbol{k}_1-\boldsymbol{k}_2)\cdot\boldsymbol{x}+\phi)]$$

複素表示を使う方法

$E_O = E_1 e^{i(\boldsymbol{k}_1\cdot\boldsymbol{x}-\omega t+\phi)}$　$E_R = E_2 e^{i(\boldsymbol{k}_2\cdot\boldsymbol{x}-\omega t)}$

$$|E_O+E_R|^2 = (E_1 e^{i(\boldsymbol{k}_1\cdot\boldsymbol{x}-\omega t+\phi)}+E_2 e^{i(\boldsymbol{k}_2\cdot\boldsymbol{x}-\omega t)})(E_1 e^{-i(\boldsymbol{k}_1\cdot\boldsymbol{x}-\omega t+\phi)}+E_2 e^{-i(\boldsymbol{k}_2\cdot\boldsymbol{x}-\omega t)}))$$

$$= E_1^{\ 2}+E_2^{\ 2}+E_1E_2 e^{i[(\boldsymbol{k}_1-\boldsymbol{k}_2)\cdot\boldsymbol{x}+\phi]}+E_1E_2 e^{-i[(\boldsymbol{k}_1-\boldsymbol{k}_2)\cdot\boldsymbol{x}+\phi]}$$

$$= E_1^{\ 2}+E_2^{\ 2}+2E_1E_2\cos[(\boldsymbol{k}_1-\boldsymbol{k}_2)\cdot\boldsymbol{x}+\phi)]$$

波数空間に波数ベクトル$\pm(\boldsymbol{k}_1-\boldsymbol{k}_2)$を記録

$E_R = E_2 e^{i(\boldsymbol{k}_2\cdot\boldsymbol{x}-\omega t)}$を照射

$$E_R|E_O+E_R|^2 = E_2 e^{i(\boldsymbol{k}_2\cdot\boldsymbol{x}-\omega t)}(E_1^{\ 2}+E_2^{\ 2}+E_1E_2 e^{i[(\boldsymbol{k}_1-\boldsymbol{k}_2)\cdot\boldsymbol{x}+\phi]}+E_1E_2 e^{-i[(\boldsymbol{k}_1-\boldsymbol{k}_2)\cdot\boldsymbol{x}+\phi]})$$

$$\to e^{i(\boldsymbol{k}_1\cdot\boldsymbol{x}-\omega t+\phi)},\quad e^{i[(2\boldsymbol{k}_2-\boldsymbol{k}_1)\cdot\boldsymbol{x}-\omega t-\phi]}$$

ホログラフィーのより詳しい解析

物体イメージ$f(x,y)$

$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} f(x,y)e^{-i(k_x x+k_y y)}dxdy = F(k_x,k_y)$**遠方での回折像**

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} f(x_0,y_0)e^{i[k_x(x-x_0)+k_y(y-y_0)]}dx_0dy_0 = e^{i(k_x x+k_y y)}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} f(x_0,y_0)e^{-i(k_x x_0+k_y y_0)}dx_0dy_0 = e^{i(k_x x+k_y y)}F(k_x,k_y)$$

時間領域ホログラフィー

信号光 $E_S = E_0(t+T)e^{i\omega_0(t+T)+i\phi}$ at $t=-T$　書き込み光　$E_R(t)=E_0(t)e^{i\omega_0 t}$ at $t=0$

$$E(\omega)=\int(E_S+E_R)e^{-i\omega t}dt=\int[E_0(t+T)e^{i\omega_0(t+T)+i\phi}+E_0(t)e^{i\omega_0 t}]e^{-i\omega t}dt$$

$$=\int[e^{i\omega T+i\phi}E_0(t+T)e^{-i(\omega-\omega_0)(t+T)}+E_0(t)e^{-i(\omega-\omega_0)t}]dt$$

$=e^{i\omega T+i\phi}E_0(\omega-\omega_0)+E_0(\omega-\omega_0)$　これに比例する分極が発生

周波数領域にpopulation分布として時間間隔$T(=0-(-T))$を記録

$$|E(\omega)|^2=|E_0(\omega-\omega_0)|^2(2+e^{i\omega T+i\phi}+e^{-i\omega T-i\phi})=|E_0(\omega-\omega_0)|^2[2+2\cos(\omega T+\phi)]$$

読み出し光　$E_R(t)=E_0(t-\tau)e^{i\omega_0(t-\tau)}$ at $t=\tau \xrightarrow{\text{F.T.}} e^{-i\omega\tau}E_0(\omega-\omega_0)$

$$\because \int[E_0(t-\tau)e^{i\omega_0(t-\tau)}]e^{-i\omega t}dt=e^{-i\omega\tau}\int E_0(t-\tau)e^{-i(\omega-\omega_0)(t-\tau)}dt=e^{-i\omega\tau}E_0(\omega-\omega_0)$$

$$e^{-i\omega\tau}E_0(\omega-\omega_0)|E(\omega)|^2=E_0(\omega-\omega_0)|E_0(\omega-\omega_0)|^2(2e^{-i\omega\tau}+e^{i\omega(T-\tau)+i\phi}+e^{-i\omega(T+\tau)-i\phi})$$

第３項　$E_0(\omega-\omega_0)|E_0(\omega-\omega_0)|^2=E'(\omega-\omega_0)$とおく

$$\frac{1}{2\pi}\int E'(\omega-\omega_0)e^{-i\omega(T+\tau)-i\phi}e^{i\omega t}d\omega=\frac{1}{2\pi}e^{i\omega_0(t-T-\tau)-i\phi}\int E'(\omega-\omega_0)e^{i(\omega-\omega_0)(t-T-\tau)}d(\omega-\omega_0)$$

$=E'(t-T-\tau)e^{i\omega_0(t-T-\tau)-i\phi}$　エコー　at $t=\tau+T$

第２項

$$\frac{1}{2\pi}\int E'(\omega-\omega_0)e^{i\omega(T-\tau)+i\phi}e^{i\omega t}d\omega=\frac{1}{2\pi}e^{i\omega_0(t+T-\tau)+i\phi}\int E'(\omega-\omega_0)e^{i(\omega-\omega_0)(t+T-\tau)}d(\omega-\omega_0)$$

$=E'(t+T-\tau)e^{i\omega_0(t+T-\tau)+i\phi}$　?　at $t=\tau-T$