

誘電率の分散の量子論 (半古典論)

物質 - 量子力学 光 - 古典的電磁波

物質: 原子(分子)の集団 原子(分子)波動関数

$$\psi_m(r, t) = e^{-i\frac{E_m t}{\hbar}} \phi_m(r) \quad E_n = \hbar\omega_n$$

ハミルトニアン $H = H_0 + H'$ H' : 光電場と物質との相互作用ハミルトニアン

$$H_0 \phi_m = E_m \phi_m$$

原子の双極子モーメント

$$\mathbf{M} = -\sum_j e \mathbf{r}_j \quad \mathbf{r}_j: j\text{番目の電子の座標}$$

$$H' = -\mathbf{M} \cdot \mathbf{E} = -\mathbf{M} \cdot \mathbf{E}_0 \cos \omega t = -\mu E_0 \cos \omega t \quad \mu: \mathbf{M} \text{ の } \mathbf{E} \text{ 方向成分}$$

光の波長 \gg 原子のサイズ \rightarrow 原子内で光電場は一定 \Rightarrow 電気双極子近似

$$\mathbf{M}_{mn} = \langle \phi_m | \mathbf{M} | \phi_n \rangle \text{ と } E_0 \text{ のなす角を } \theta$$

$$\mu_{mn} = \langle \phi_m | M \cos \theta | \phi_n \rangle = \langle \phi_m | M_x | \phi_n \rangle$$

$$\text{等方的物質の場合} \rightarrow \langle m | M_x | n \rangle = \langle m | M_y | n \rangle = \langle m | M_z | n \rangle$$

$$\therefore |\mathbf{M}_{mn}|^2 = 3 |\mu_{mn}|^2$$

$$\Psi(r, t) = \sum_n C_n'(t) \psi_n(r, t) = \sum_n C_n'(t) e^{-i\frac{E_n t}{\hbar}} \phi_n(r) = \sum_n C_n(t) \phi_n(r) \quad C_n(t) = C_n'(t) e^{-i\frac{E_n t}{\hbar}}$$

$$H\Psi = i\hbar \frac{\partial \Psi}{\partial t}$$

$$(H_0 + H')(\sum_n C_n \phi_n) = i\hbar \frac{\partial}{\partial t} (\sum_n C_n \phi_n)$$

$$\langle \phi_m | H_0 + H' | \sum_n C_n \phi_n \rangle = \langle \phi_m | i\hbar \sum_n \frac{dC_n}{dt} \phi_n \rangle$$

$$C_m E_m + \sum_n C_n \langle \phi_m | H' | \phi_n \rangle = i\hbar \sum_n \frac{dC_n}{dt} \langle \phi_m | \phi_n \rangle$$

$$i\hbar \frac{dC_m}{dt} = E_m C_m + \sum_n C_n H'_{mn}$$

$$i\hbar \frac{dC_m(t)}{dt} = E_m C_m(t) + \sum_n C_n(t) (-\mu_{mn}) E_0 \cos \omega t$$

二準位系で、原子など中心対称性(反転対称性)がある場合を考える

ϕ_n : even $\mathbf{r} \rightarrow -\mathbf{r}$ で波動関数が不変 $odd \quad \mathbf{r} \rightarrow -\mathbf{r}$ で符号のみ反転

$$\therefore \mu_{ii} = 0$$

$$\left\{ \begin{array}{l} i\hbar \frac{dC_1}{dt} = E_1 C_1 - (\mu_{12} E_0 \cos \omega t) C_2 \\ i\hbar \frac{dC_2}{dt} = E_2 C_2 - (\mu_{21} E_0 \cos \omega t) C_1 \end{array} \right\}$$

$$\left\{ \begin{array}{l} \frac{dC_1}{dt} = -i\omega_1 C_1 + i\left(\frac{\mu_{12} E_0}{\hbar} \cos \omega t\right) C_2 = -i\omega_1 C_1 + i(\xi \cos \omega t) C_2 = -i\omega_1 C_1 + i\eta(t) C_2 \\ \frac{dC_2}{dt} = -i\omega_2 C_2 + i\left(\frac{\mu_{21} E_0}{\hbar} \cos \omega t\right) C_1 = -i\omega_2 C_2 + i(\xi^* \cos \omega t) C_1 = -i\omega_2 C_2 + i\eta^*(t) C_1 \end{array} \right\}$$

$$\xi = \frac{\mu_{12} E_0}{\hbar} \quad \xi^* = \frac{\mu_{21} E_0}{\hbar} \quad \eta(t) = \xi \cos \omega t$$

密度行列

$$\rho_{11} = C_1 C_1^* \quad \rho_{12} = C_1 C_2^*$$

$$\rho_{22} = C_2 C_2^* \quad \rho_{21} = C_2 C_1^*$$

$$\frac{d\rho_{11}}{dt} = \frac{dC_1}{dt} C_1^* + C_1 \frac{dC_1^*}{dt} = (-i\omega_1 C_1 + i\eta C_2) C_1^* + C_1 (i\omega_1 C_1^* - i\eta^* C_2^*) = i(\eta \rho_{21} - \eta^* \rho_{12})$$

$$\frac{d\rho_{22}}{dt} = -\frac{d\rho_{11}}{dt} \quad \because \rho_{22} = 1 - \rho_{11}$$

$$\begin{aligned} \frac{d\rho_{12}}{dt} &= \frac{dC_1}{dt} C_2^* + C_1 \frac{dC_2^*}{dt} = (-i\omega_1 C_1 + i\eta C_2) C_2^* + C_1 (i\omega_2 C_2^* - i\eta^* C_1^*) \\ &= i(\omega_2 - \omega_1) C_1 C_2^* + i\eta(C_2 C_2^* - C_1 C_1^*) \\ &= i\omega_0 \rho_{12} + i\eta(\rho_{22} - \rho_{11}) \quad \omega_0 = \omega_2 - \omega_1 \end{aligned}$$

$$\frac{d\rho_{21}}{dt} = \frac{d\rho_{12}^*}{dt}$$

$$\frac{d\rho_{11}}{dt} = i(\eta \rho_{21} - \eta^* \rho_{12}) + \gamma_1 \rho_{22}$$

$$\frac{d\rho_{22}}{dt} = -i(\eta \rho_{21} - \eta^* \rho_{12}) - \gamma_1 \rho_{22}$$

$$\frac{d\rho_{12}}{dt} = i\omega_0 \rho_{12} + i\eta(\rho_{22} - \rho_{11}) - \gamma_2 \rho_{12}$$

$$\frac{d\rho_{21}}{dt} = -i\omega_0 \rho_{21} - i\eta^*(\rho_{22} - \rho_{11}) - \gamma_2 \rho_{21}$$

ρ_{12} の解

$$\rho_{22} = 0, \rho_{11} = 1$$

$$\frac{d\rho_{12}}{dt} - i\omega_0 \rho_{12} + \gamma_2 \rho_{12} = -i\eta = -i\xi \cos \omega t = -i\frac{\xi}{2}(e^{i\omega t} + e^{-i\omega t})$$

$$\text{定常解 } \rho_{12} = ae^{i\omega t} + be^{-i\omega t}$$

$$[i(\omega - \omega_0) + \gamma_2]ae^{i\omega t} + [-i(\omega + \omega_0) + \gamma_2]be^{-i\omega t} = -i\frac{\xi}{2}(e^{i\omega t} + e^{-i\omega t})$$

$$\rho_{12} = \frac{-i\frac{\xi}{2}e^{i\omega t}}{i(\omega - \omega_0) + \gamma_2} + \frac{-i\frac{\xi}{2}e^{-i\omega t}}{-i(\omega + \omega_0) + \gamma_2} = \frac{\xi}{2} \left[\frac{e^{i\omega t}}{(\omega_0 - \omega) + i\gamma_2} + \frac{e^{-i\omega t}}{(\omega_0 + \omega) + i\gamma_2} \right]$$

巨視的分極

$$P(t) = \frac{N}{V} \langle \Psi | \mu | \Psi \rangle = \frac{N}{V} \langle C_1 \phi_1 + C_2 \phi_2 | \mu | C_1 \phi_1 + C_2 \phi_2 \rangle = \frac{N}{V} (C_1^* C_2 \mu_{12} + C_2^* C_1 \mu_{21}) = \frac{N}{V} (\rho_{21} \mu_{12} + \rho_{12} \mu_{21})$$

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

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

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

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

$$\begin{aligned} \chi(\omega) &= \frac{N}{V} \frac{|\mu_{12}|^2}{\varepsilon_0 \hbar} \left(\frac{1}{\omega_0 - \omega - i\gamma_2} + \frac{1}{\omega_0 + \omega + i\gamma_2} \right) = \frac{N}{V} \frac{|\mu_{12}|^2}{\varepsilon_0 \hbar} \frac{2\omega_0}{\omega_0^2 - (\omega + i\gamma_2)^2} \quad \gamma_2 = \Gamma_0/2 \\ &= \frac{N}{V} \frac{|\mu_{12}|^2}{\varepsilon_0 \hbar} \frac{2\omega_0}{\omega_0^2 - \omega^2 + \Gamma_0^2/4 - i\omega\Gamma_0} \\ &= \frac{1}{\varepsilon_0} \frac{N e^2}{V m} \frac{\frac{2m\omega_0 |\mu_{12}|^2}{\hbar e^2}}{\omega_0^2 - \omega^2 + \Gamma_0^2/4 - i\omega\Gamma_0} \rightarrow \frac{1}{\varepsilon_0} \frac{N e^2}{V m} \sum_j \frac{f_{gj}}{\omega_{gj}^2 - \omega^2 - i\omega\Gamma_{gj}} \\ &\quad \omega_0^2 + \Gamma_0^2/4 \rightarrow \omega_0^2 \quad \omega_0 \rightarrow \omega_{gj} \quad \Gamma_0 \rightarrow \Gamma_{gj} \quad f_{gj} = \frac{2m\omega_{gj} |\mu_{gj}|^2}{\hbar e^2} \end{aligned}$$

f_{gj} : 振動子強度

振動子強度の総和則

$$\chi(\omega) = \frac{1}{\varepsilon_0} \frac{N e^2}{V m} \sum_j \frac{f_{gj}}{\omega_{gj}^2 - \omega^2 - i\omega\Gamma_{gj}}$$

$\omega \rightarrow \infty (\omega \gg \omega_{gj})$ 自由電子の振る舞いに近づく

$$\chi(\omega) \rightarrow \frac{1}{\varepsilon_0} \frac{N e^2}{V m} \left(-\frac{1}{\omega^2} \sum_j f_{gj} \right) \dots (A)$$

自由電子の感受率

$$m \frac{d^2 x}{dt^2} = -eE \quad x = \frac{eE}{m\omega^2}$$

$$p = -ex = -\frac{e^2 E}{m\omega^2}$$

$$P = \frac{N}{V} p = -\frac{N e^2 E}{V m\omega^2} = \varepsilon_0 \chi E$$

$$\chi = -\frac{1}{\varepsilon_0} \frac{N e^2}{V m\omega^2} \dots (B)$$

(A)と(B)を比較して

$$\therefore \sum_j f_{gj} = 1 \quad 1\text{電子系} \quad N\text{電子系} \sum_j f_{gj} = N \quad \text{総和則}$$

水素原子の許容遷移 f_{gj}

1S - 2P	121.567nm	0.4162
1S - 3P	102.572nm	0.07910
1S - 4P	97.254nm	0.02899

$$A_{2P-1S} = 6.26 \times 10^8 \text{s}^{-1} = \gamma_1 = 2\gamma_2$$

$$\begin{aligned}\frac{d\rho_{11}}{dt} &= i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_1\rho_{11} + \Gamma_2\rho_{22} \\ \frac{d\rho_{22}}{dt} &= -i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_2\rho_{22} + \Gamma_1\rho_{11} \\ \frac{d\rho_{12}}{dt} &= i\omega_0\rho_{12} + i\eta(\rho_{22} - \rho_{11}) - \gamma_2\rho_{12} \\ \frac{d\rho_{21}}{dt} &= -i\omega_0\rho_{21} - i\eta^*(\rho_{22} - \rho_{11}) - \gamma_2\rho_{21}\end{aligned}$$

$$T \rightarrow 0 \quad \Gamma_2 \rightarrow \gamma_1 \quad \Gamma_1 \rightarrow 0$$

$$\begin{aligned}\frac{d\rho_{11}}{dt} &= i(\eta\rho_{21} - \eta^*\rho_{12}) + \gamma_1\rho_{22} \\ \frac{d\rho_{22}}{dt} &= -i(\eta\rho_{21} - \eta^*\rho_{12}) - \gamma_1\rho_{22} \\ \frac{d\rho_{12}}{dt} &= i\omega_0\rho_{12} + i\eta(\rho_{22} - \rho_{11}) - \gamma_2\rho_{12} \\ \frac{d\rho_{21}}{dt} &= -i\omega_0\rho_{21} - i\eta^*(\rho_{22} - \rho_{11}) - \gamma_2\rho_{21}\end{aligned}$$

$$\begin{aligned}\frac{d\rho_{11}}{dt} &= \frac{i}{2}(\xi\rho_{21} - \xi^*\rho_{12})(e^{i\omega t} + e^{-i\omega t}) + \gamma_1\rho_{22} \\ \frac{d\rho_{22}}{dt} &= -\frac{i}{2}(\xi\rho_{21} - \xi^*\rho_{12})(e^{i\omega t} + e^{-i\omega t}) - \gamma_1\rho_{22} \\ \frac{d\rho_{12}}{dt} &= i\omega_0\rho_{12} + \frac{i}{2}\xi(\rho_{22} - \rho_{11})(e^{i\omega t} + e^{-i\omega t}) - \gamma_2\rho_{12} \\ \frac{d\rho_{21}}{dt} &= -i\omega_0\rho_{21} - \frac{i}{2}\xi^*(\rho_{22} - \rho_{11})(e^{i\omega t} + e^{-i\omega t}) - \gamma_2\rho_{21}\end{aligned}$$

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

$$\begin{aligned}\frac{d\tilde{\rho}_{12}}{dt} &= i\omega_0\rho_{12}e^{-i\omega t} + \frac{i}{2}\xi(\rho_{22} - \rho_{11})(1 + e^{-i2\omega t}) - \gamma_2\rho_{12}e^{-i\omega t} - i\omega\tilde{\rho}_{12} \\ &\cong i(\omega_0 - \omega)\tilde{\rho}_{12} + \frac{i}{2}\xi(\rho_{22} - \rho_{11}) - \gamma_2\tilde{\rho}_{12} \quad \text{回転波近似}\end{aligned}$$

$$\begin{aligned}\frac{d\rho_{22}}{dt} &= -\frac{i}{2}(\xi\tilde{\rho}_{21}e^{-i\omega t} - \xi^*\tilde{\rho}_{12}e^{i\omega t})(e^{i\omega t} + e^{-i\omega t}) - \gamma_1\rho_{22} \\ &\cong -\frac{i}{2}(\xi\tilde{\rho}_{21} - \xi^*\tilde{\rho}_{12}) - \gamma_1\rho_{22}\end{aligned}$$

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

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

$$\rho_{11} = \rho_{11}^{(0)}e^{\lambda t} \quad \rho_{22} = \rho_{22}^{(0)}e^{\lambda t} \quad \tilde{\rho}_{12} = \tilde{\rho}_{12}^{(0)}e^{\lambda t} \quad \tilde{\rho}_{21} = \tilde{\rho}_{21}^{(0)}e^{\lambda t}$$

$$\rho_{11} = \rho_{11}^{(0)} e^{\lambda t} \quad \rho_{22} = \rho_{22}^{(0)} e^{\lambda t} \quad \tilde{\rho}_{12} = \tilde{\rho}_{12}^{(0)} e^{\lambda t} \quad \tilde{\rho}_{21} = \tilde{\rho}_{21}^{(0)} e^{\lambda t}$$

$$\begin{pmatrix} -\lambda & \gamma_1 & -\frac{i}{2}\xi^* & \frac{i}{2}\xi \\ 0 & -\lambda - \gamma_1 & \frac{i}{2}\xi^* & -\frac{i}{2}\xi \\ -\frac{i}{2}\xi & \frac{i}{2}\xi & -\lambda + i(\omega_0 - \omega) - \gamma_2 & 0 \\ \frac{i}{2}\xi^* & -\frac{i}{2}\xi^* & 0 & -\lambda - i(\omega_0 - \omega) - \gamma_2 \end{pmatrix} \begin{pmatrix} \rho_{11}^{(0)} \\ \rho_{22}^{(0)} \\ \tilde{\rho}_{12}^{(0)} \\ \tilde{\rho}_{21}^{(0)} \end{pmatrix} = 0$$

$$\gamma_1 = \gamma_2 = 0 \text{ のとき } \lambda^2 [\lambda^2 + |\xi|^2 + (\omega_0 - \omega)^2] = 0 \quad \lambda = 0, \pm i\sqrt{|\xi|^2 + (\omega_0 - \omega)^2} (\equiv \pm \Omega i)$$

$$\rho_{12}(t) = \tilde{\rho}_{12}(t) e^{i\omega t} = (Ae^{i\Omega t} + Be^{-i\Omega t} + C)e^{i\omega t}$$

$$\rho_{22}(t) = \alpha e^{i\Omega t} + \beta e^{-i\Omega t} + \gamma$$

$$\text{初期条件 } \rho_{12}(0) = 0 \quad \rho_{11}(0) = 1 \quad \rho_{22}(0) = 0 \quad \left(\frac{d\tilde{\rho}_{12}}{dt} \right)_{t=0} = -\frac{i}{2}\xi \quad \left(\frac{d\rho_{22}}{dt} \right)_{t=0} = 0$$

$$\left(\frac{d^2 \tilde{\rho}_{12}}{dt^2} \right)_{t=0} = i(\omega_0 - \omega) \frac{d\tilde{\rho}_{12}}{dt} + \frac{i}{2}\xi \left(\frac{d\rho_{22}}{dt} - \frac{d\rho_{11}}{dt} \right) = \frac{\xi}{2}(\omega_0 - \omega)$$

$$\left(\frac{d^2 \rho_{22}}{dt^2} \right)_{t=0} = -\frac{i}{2} \left(\xi \frac{d\tilde{\rho}_{21}}{dt} - \xi^* \frac{d\tilde{\rho}_{12}}{dt} \right) = -\frac{i}{2} \left[\xi \frac{i}{2}\xi^* - \xi^* \left(-\frac{i}{2}\xi \right) \right] = -\frac{i}{2} \cdot i |\xi|^2 = \frac{|\xi|^2}{2}$$

$$A + B + C = 0 \quad i\Omega(A - B) = -\frac{i}{2}\xi \quad -\Omega^2(A + B) = \frac{\xi}{2}(\omega_0 - \omega)$$

$$\alpha + \beta + \gamma = 0 \quad i\Omega(\alpha - \beta) = 0 \quad -\Omega^2(\alpha + \beta) = \frac{|\xi|^2}{2}$$

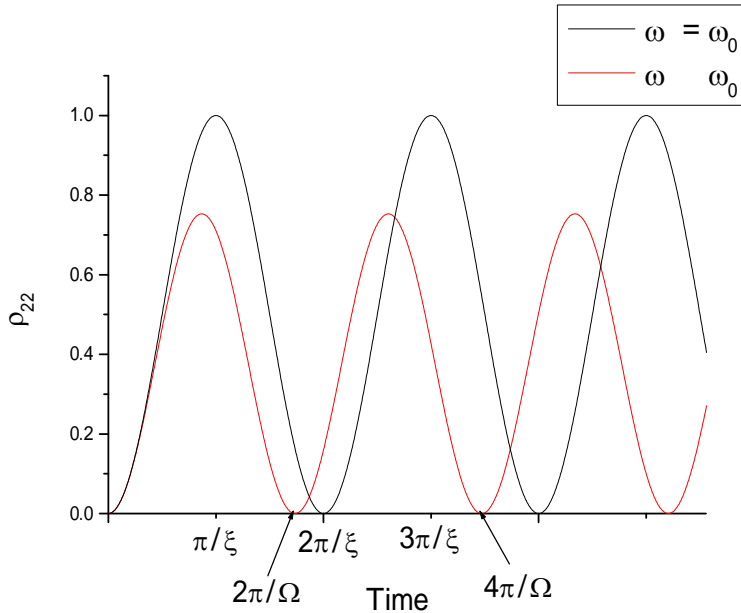
$$A = -\frac{\xi}{4\Omega^2} [\Omega + (\omega_0 - \omega)] \quad B = \frac{\xi}{4\Omega^2} [\Omega - (\omega_0 - \omega)] \quad C = \frac{\xi}{2\Omega^2} (\omega_0 - \omega)$$

$$\alpha = -\frac{|\xi|^2}{4\Omega^2} \quad \beta = -\frac{|\xi|^2}{4\Omega^2} \quad \gamma = \frac{|\xi|^2}{2\Omega^2}$$

$$\rho_{22}(t) = -\frac{|\xi|^2}{4\Omega^2} (e^{i\Omega t} + e^{-i\Omega t} - 2) = -\frac{|\xi|^2}{4\Omega^2} (2 \cos \Omega t - 2) = \frac{|\xi|^2}{\Omega^2} \frac{1 - \cos \Omega t}{2} = \frac{|\xi|^2}{\Omega^2} \sin^2 \frac{\Omega}{2} t$$

$$\Omega = \sqrt{|\xi|^2 + (\omega_0 - \omega)^2} : \text{Rabi周波数}$$

$$\text{For } \omega = \omega_0, \quad \Omega = |\xi| \quad \rho_{22}(t) = \sin^2 \frac{|\xi|}{2} t$$



振動が観測できるためには $\omega = \omega_0$ で $|\xi| \gg 3\gamma_2$

水素原子 1s 2p $3\gamma_2 \sim 10^9 \text{ s}^{-1}$ $|\xi| = 3\gamma_2$ $E_0 \sim 10^5 \text{ V/m}$ $I \sim 10^3 \text{ W/cm}^2$

Na の D 線 589nm 自然幅 $\sim 10 \text{ MHz} = 10^7 \text{ s}^{-1}$ $E_0 \sim 10^3 \text{ V/m}$ $I \sim 0.1 \text{ W/cm}^2$

地上の太陽光(真夏、晴天) $I \approx 1 \text{ kW/m}^2 = 0.1 \text{ W/cm}^2$

水素原子 1s 2p $|\xi| = \omega_0$ のためには、 $E_0 = 3 \times 10^{11} \text{ V/m}$ の光電場が必要

分極は？

$$\rho_{12}(t) = \tilde{\rho}_{12}(t)e^{i\omega t} = (Ae^{i\Omega t} + Be^{-i\Omega t} + C)e^{i\omega t}$$

$$A = -\frac{\xi}{4\Omega^2} [\Omega + (\omega_0 - \omega)] \quad B = \frac{\xi}{4\Omega^2} [\Omega - (\omega_0 - \omega)] \quad C = \frac{\xi}{2\Omega^2} (\omega_0 - \omega)$$

$$\rho_{12}(t) = \left\{ \frac{\xi}{4\Omega^2} [-\Omega - (\omega_0 - \omega)]e^{i\Omega t} + \frac{\xi}{4\Omega^2} [\Omega - (\omega_0 - \omega)]e^{-i\Omega t} + \frac{\xi}{2\Omega^2} (\omega_0 - \omega) \right\} e^{i\omega t}$$

$$= \left\{ \frac{\xi}{4\Omega} [-e^{i\Omega t} + e^{-i\Omega t}] - \frac{\xi(\omega_0 - \omega)}{4\Omega^2} (e^{i\Omega t} + e^{-i\Omega t}) + \frac{\xi(\omega_0 - \omega)}{2\Omega^2} \right\} e^{i\omega t}$$

$$= \left\{ \frac{\xi}{2\Omega} (-i \sin \Omega t) + \frac{\xi(\omega_0 - \omega)}{2\Omega^2} (1 - \cos \Omega t) \right\} e^{i\omega t}$$

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

$$= \frac{N}{V} \frac{|\mu_{12}|^2 E_0}{\Omega \hbar} \left\{ \left[\left(-\frac{i}{2} \sin \Omega t \right) e^{i\omega t} + \left(\frac{i}{2} \sin \Omega t \right) e^{-i\omega t} \right] + \frac{(\omega_0 - \omega)}{2\Omega} (1 - \cos \Omega t) (e^{i\omega t} + e^{-i\omega t}) \right\}$$

$$= \frac{N}{V} \frac{|\mu_{12}|^2 E_0}{\Omega \hbar} \left\{ \sin \Omega t \sin \omega t + \frac{\omega_0 - \omega}{\Omega} (1 - \cos \Omega t) \cos \omega t \right\}$$

$$\text{For } \omega = \omega_0, \quad P(t) = \frac{N}{V} \frac{|\mu_{12}|^2 E_0}{\Omega \hbar} \sin \xi t \sin \omega t$$

2準位系、中心対称、回転波近似、 $\gamma_1, \gamma_2 = 0$ のもとで正しい式

$$\rho_{22}(t) = \frac{|\xi|^2}{\Omega^2} \sin^2 \frac{\Omega}{2} t$$

$$\rho_{22}(t) \rightarrow |\xi|^2 \frac{\sin^2 \frac{(\omega_0 - \omega)t}{2}}{(\omega_0 - \omega)^2} \quad (\omega_0 - \omega)^2 \gg |\xi|^2$$

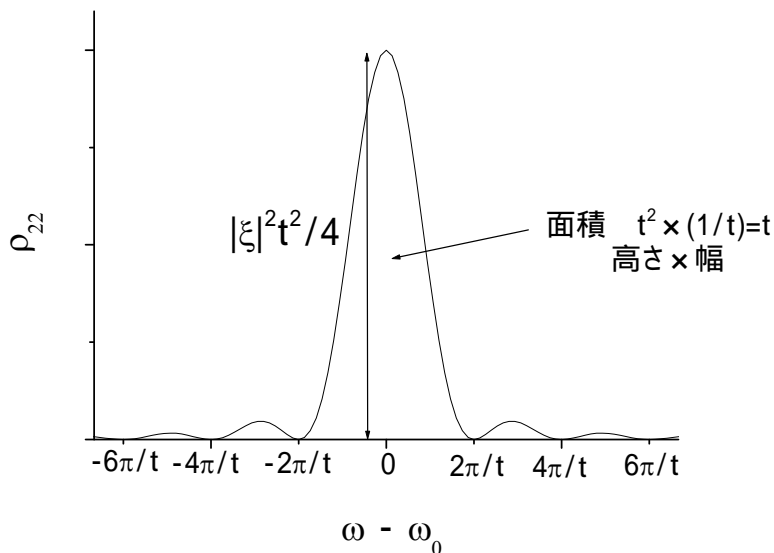
$\Omega = \sqrt{|\xi|^2 + (\omega_0 - \omega)^2}$: Rabi周波数

$\Omega t \ll 1$ ($\rho_{22} \ll 1$)の条件で、

$\omega \rightarrow \omega_0$ のとき、

$$\rho_{22}(t) \rightarrow \frac{|\xi|^2}{4} t^2$$

$\omega = \omega_0$ のとき、 $(\omega_0 - \omega)^2 \gg |\xi|^2$ が成り立たないが、 $\rho_{22}(t) = \sin^2 \frac{|\xi|}{2} t \rightarrow \frac{|\xi|^2}{4} t^2$ でOK



$$\rho_{22}(t) = \frac{|\xi|^2}{2} t \frac{\sin^2((\omega_0 - \omega)t/2)}{(\omega_0 - \omega)^2 t/2} \xrightarrow{t \rightarrow \infty} \frac{1}{\pi} \lim_{t \rightarrow \infty} \frac{\sin^2((\omega_0 - \omega)t/2)}{(\omega_0 - \omega)^2 t/2} \rightarrow \delta(\omega_0 - \omega) \text{より}$$

$$\rho_{22}(t) = \frac{\pi |\xi|^2}{2} t \delta(\omega_0 - \omega) \quad \text{遷移確率 } w_{12} = \frac{d\rho_{22}}{dt} = \frac{\pi}{2\hbar^2} |\mu_{12} E_0|^2 \delta(\omega_0 - \omega) \quad \text{Fermiの黄金律}$$

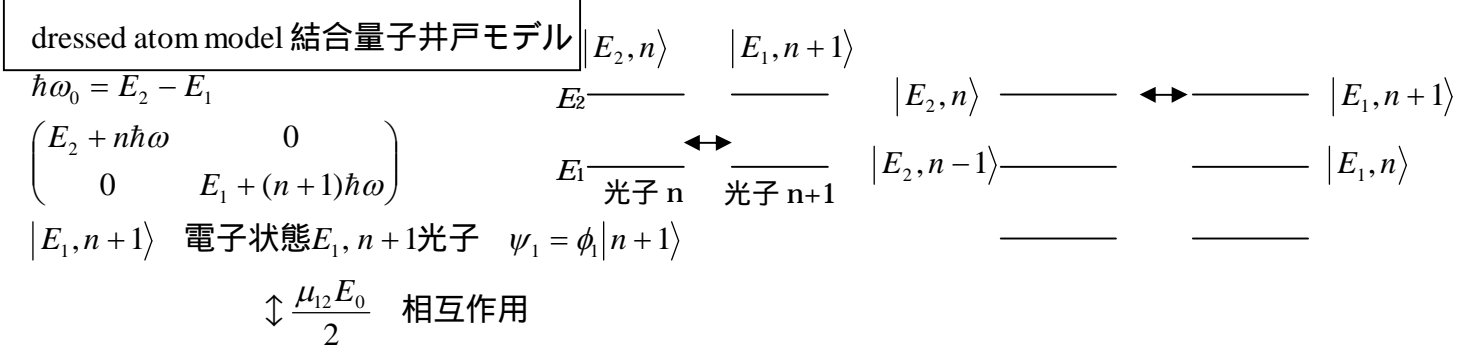
$$\frac{\varepsilon_0}{2} E_0^2 = \int U(\omega) d\omega$$

$$\rho_{22}(t) = \frac{|\xi|^2}{4} \frac{\sin^2\left(\frac{\omega_0 - \omega}{2} t\right)}{\left(\frac{\omega_0 - \omega}{2}\right)^2} = \frac{1}{2\varepsilon_0} \frac{|\mu_{12}|^2}{\hbar^2} \frac{\varepsilon_0 E_0^2}{2} \frac{\sin^2\left(\frac{\omega_0 - \omega}{2} t\right)}{\left(\frac{\omega_0 - \omega}{2}\right)^2} = \frac{1}{2\varepsilon_0} \frac{|\mu_{12}|^2}{\hbar^2} \int d\omega U(\omega) \frac{\sin^2\left(\frac{\omega_0 - \omega}{2} t\right)}{\left(\frac{\omega_0 - \omega}{2}\right)^2}$$

$$= \frac{1}{2\varepsilon_0} \frac{|\mu_{12}|^2}{\hbar^2} U(\omega_0) \int d\omega \frac{\sin^2\left(\frac{\omega_0 - \omega}{2} t\right)}{\left(\frac{\omega_0 - \omega}{2}\right)^2} = \frac{1}{2\varepsilon_0} \frac{|\mu_{12}|^2}{\hbar^2} U(\omega_0) \cdot 2t \int_{-\infty}^{\infty} \frac{\sin^2 x}{x^2} dx = \frac{1}{2\varepsilon_0} \frac{|\mu_{12}|^2}{\hbar^2} U(\omega_0) \cdot 2\pi$$

$$\text{遷移確率 } w_{12} = \frac{d\rho_{22}(t)}{dt} = \frac{\pi |\mu_{12}|^2}{\varepsilon_0 \hbar^2} U(\omega_0) \equiv B_{12} U(\omega_0) \quad B_{12} = \frac{\pi |\mu_{12}|^2}{\varepsilon_0 \hbar^2} \left[\frac{\text{m}^3}{\text{J} \cdot \text{s}^2} \right] \text{ EinsteinのB係数 誘導吸収・放出}$$

Rabi 振動を理解するための



$|E_2, n\rangle$ 電子状態 E_2, n 光子 $\psi_2 = \phi_2 |n\rangle$

$$\begin{vmatrix} E_2 + n\hbar\omega - \lambda & \frac{\mu_{12} E_0}{2} \\ \frac{\mu_{12}^* E_0}{2} & E_1 + (n+1)\hbar\omega - \lambda \end{vmatrix} = (E_2 + n\hbar\omega - \lambda)(E_1 + (n+1)\hbar\omega - \lambda) - \frac{|\mu_{12} E_0|^2}{4}$$

$$= \lambda^2 - [E_1 + E_2 + (2n+1)\hbar\omega]\lambda + (E_2 + n\hbar\omega)(E_1 + (n+1)\hbar\omega) - \frac{|\mu_{12} E_0|^2}{4} = 0$$

固有値 $\lambda = \frac{1}{2} [E_1 + E_2 + (2n+1)\hbar\omega \pm \sqrt{(E_1 - E_2 + \hbar\omega)^2 + |\mu_{12} E_0|^2}]$

$\hbar\omega_0 = E_2 - E_1$ $\hbar\Delta = E_1 + E_2 + (2n+1)\hbar\omega$ $\delta = \omega_0 - \omega$ $\xi = \frac{\mu_{12} E_0}{\hbar}$ $\Omega = \sqrt{\delta^2 + |\xi|^2}$

$\lambda_{\pm} = \frac{1}{2} \hbar(\Delta \pm \Omega)$

固有状態 Ψ_+, Ψ_-

$\lambda = \lambda_+ \begin{pmatrix} \frac{1}{2} \hbar(\delta - \Omega) & \frac{1}{2} \hbar\xi \\ \frac{1}{2} \hbar\xi^* & \frac{1}{2} \hbar(-\delta - \Omega) \end{pmatrix} \begin{pmatrix} x_+ \\ y_+ \end{pmatrix} = 0 \quad \begin{pmatrix} x_+ \\ y_+ \end{pmatrix} = \frac{1}{\sqrt{|\xi|^2 + (\Omega - \delta)^2}} \begin{pmatrix} \xi \\ \Omega - \delta \end{pmatrix} \equiv \Psi_+$

$\lambda = \lambda_- \begin{pmatrix} \frac{1}{2} \hbar(\delta + \Omega) & \frac{1}{2} \hbar\xi \\ \frac{1}{2} \hbar\xi^* & \frac{1}{2} \hbar(-\delta + \Omega) \end{pmatrix} \begin{pmatrix} x_- \\ y_- \end{pmatrix} = 0 \quad \begin{pmatrix} x_- \\ y_- \end{pmatrix} = \frac{1}{\sqrt{|\xi|^2 + (\Omega + \delta)^2}} \begin{pmatrix} \xi \\ -\Omega - \delta \end{pmatrix} \equiv \Psi_-$

$\Psi_+ = \frac{1}{\sqrt{|\xi|^2 + (\Omega - \delta)^2}} [\xi\psi_2 + (\Omega - \delta)\psi_1]$

$\Psi_- = \frac{1}{\sqrt{|\xi|^2 + (\Omega + \delta)^2}} [\xi\psi_2 - (\Omega + \delta)\psi_1]$

任意の状態 Ψ は $\Psi_+ e^{-i\frac{\lambda_+}{\hbar}t}$, $\Psi_- e^{-i\frac{\lambda_-}{\hbar}t}$ の線形結合で表せる

$\Psi = C_1 e^{-i\frac{\lambda_+}{\hbar}t} \Psi_+ + C_2 e^{-i\frac{\lambda_-}{\hbar}t} \Psi_- = C_1 e^{-i\frac{\Delta + \Omega}{2}t} \Psi_+ + C_2 e^{-i\frac{\Delta - \Omega}{2}t} \Psi_-$
 $= e^{-i\frac{\Delta}{2}t} [C_1 e^{-i\frac{\Omega}{2}t} \Psi_+ + C_2 e^{i\frac{\Omega}{2}t} \Psi_-]$

$= e^{-i\frac{\Delta}{2}t} \left[C_1 e^{-i\frac{\Omega}{2}t} \frac{\xi\psi_2 + (\Omega - \delta)\psi_1}{\sqrt{|\xi|^2 + (\Omega - \delta)^2}} + C_2 e^{i\frac{\Omega}{2}t} \frac{\xi\psi_2 - (\Omega + \delta)\psi_1}{\sqrt{|\xi|^2 + (\Omega + \delta)^2}} \right]$

$= e^{-i\frac{\Delta}{2}t} \left[\left(\frac{\xi C_1 e^{-i\frac{\Omega}{2}t}}{\sqrt{|\xi|^2 + (\Omega - \delta)^2}} + \frac{\xi C_2 e^{i\frac{\Omega}{2}t}}{\sqrt{|\xi|^2 + (\Omega + \delta)^2}} \right) \psi_2 + \left(\frac{(\Omega - \delta) C_1 e^{-i\frac{\Omega}{2}t}}{\sqrt{|\xi|^2 + (\Omega - \delta)^2}} - \frac{(\Omega + \delta) C_2 e^{i\frac{\Omega}{2}t}}{\sqrt{|\xi|^2 + (\Omega + \delta)^2}} \right) \psi_1 \right]$

初期条件 $t = 0$ で $\Psi = \psi_1 = \phi_1 |n+1\rangle$

$$\frac{\xi C_1}{\sqrt{|\xi|^2 + (\Omega - \delta)^2}} + \frac{\xi C_2}{\sqrt{|\xi|^2 + (\Omega + \delta)^2}} = 0$$

$$\frac{(\Omega - \delta) C_1}{\sqrt{|\xi|^2 + (\Omega - \delta)^2}} - \frac{(\Omega + \delta) C_2}{\sqrt{|\xi|^2 + (\Omega + \delta)^2}} = 1$$

$$|\xi|^2 + (\Omega - \delta)^2 = |\xi|^2 + \Omega^2 - 2\Omega\delta + \delta^2 = 2\Omega^2 - 2\Omega\delta = 2\Omega(\Omega - \delta)$$

$$|\xi|^2 + (\Omega + \delta)^2 = |\xi|^2 + \Omega^2 + 2\Omega\delta + \delta^2 = 2\Omega^2 + 2\Omega\delta = 2\Omega(\Omega + \delta)$$

$$\frac{C_1}{\sqrt{\Omega - \delta}} + \frac{C_2}{\sqrt{\Omega + \delta}} = 0$$

$$\sqrt{\Omega - \delta} C_1 - \sqrt{\Omega + \delta} C_2 = \sqrt{2\Omega}$$

$$C_1 = \sqrt{\frac{\Omega - \delta}{2\Omega}} \quad C_2 = -\sqrt{\frac{\Omega + \delta}{2\Omega}}$$

$$\Psi = e^{-i\frac{\Delta}{2}t} \left[\left(\sqrt{\frac{\Omega - \delta}{2\Omega}} \frac{\xi e^{-i\frac{\Omega}{2}t}}{\sqrt{2\Omega(\Omega - \delta)}} - \sqrt{\frac{\Omega + \delta}{2\Omega}} \frac{\xi e^{i\frac{\Omega}{2}t}}{\sqrt{2\Omega(\Omega + \delta)}} \right) \psi_2 + \left(\sqrt{\frac{\Omega - \delta}{2\Omega}} \frac{(\Omega - \delta) e^{-i\frac{\Omega}{2}t}}{\sqrt{2\Omega(\Omega - \delta)}} + \sqrt{\frac{\Omega + \delta}{2\Omega}} \frac{(\Omega + \delta) e^{i\frac{\Omega}{2}t}}{\sqrt{2\Omega(\Omega + \delta)}} \right) \psi_1 \right]$$

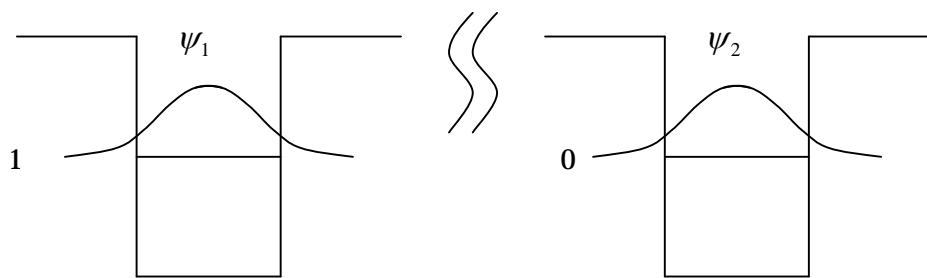
$$= e^{-i\frac{\Delta}{2}t} \left[\left(\frac{\xi e^{-i\frac{\Omega}{2}t}}{2\Omega} - \frac{\xi e^{i\frac{\Omega}{2}t}}{2\Omega} \right) \psi_2 + \left(\frac{(\Omega - \delta) e^{-i\frac{\Omega}{2}t}}{2\Omega} + \frac{(\Omega + \delta) e^{i\frac{\Omega}{2}t}}{2\Omega} \right) \psi_1 \right]$$

$$= e^{-i\frac{\Delta}{2}t} \left[\left(-i \frac{\xi}{\Omega} \sin \frac{\Omega}{2} t \right) \psi_2 + \left(\cos \frac{\Omega}{2} t + i \frac{\delta}{\Omega} \sin \frac{\Omega}{2} t \right) \psi_1 \right]$$

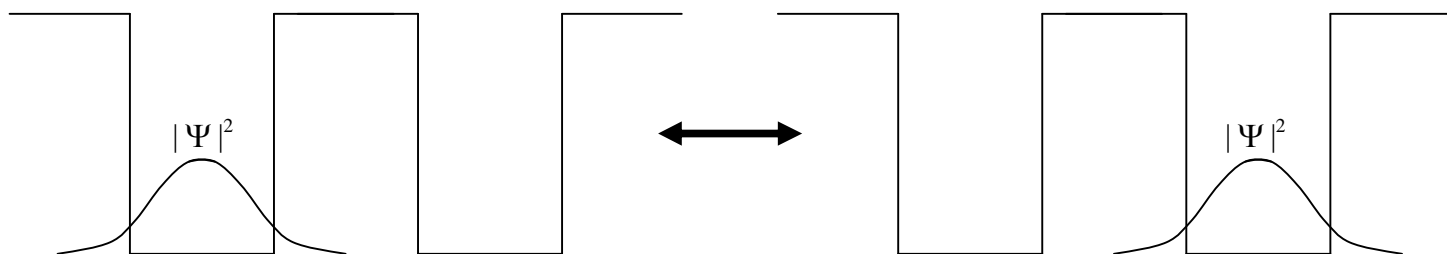
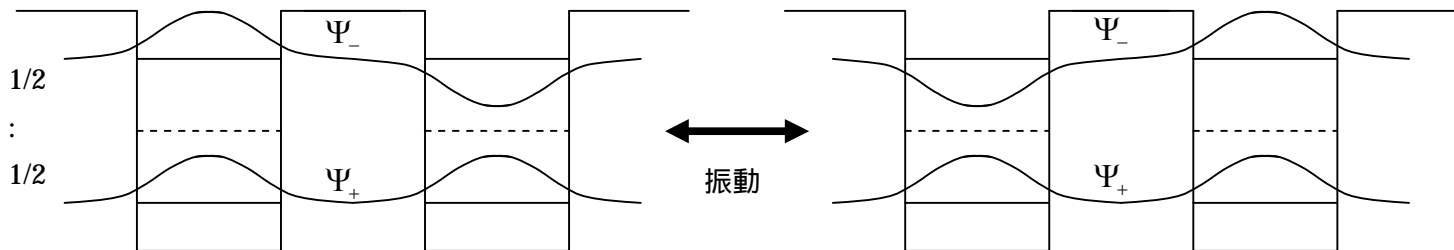
$\psi_2 = \phi_2 |n\rangle$ の確率 $\frac{|\xi|^2}{\Omega^2} \sin^2 \frac{\Omega}{2} t$ Rabi振動

共鳴のとき、 $\omega = \omega_0$ $E_1 + (n+1)\hbar\omega = E_2 + n\hbar\omega$

相互作用なし

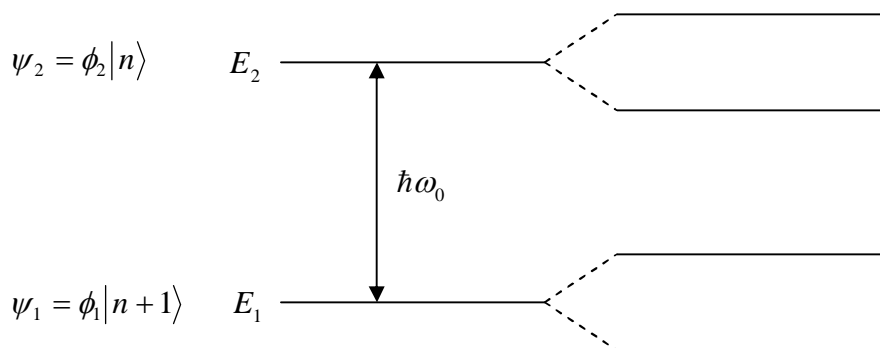


相互作用あり



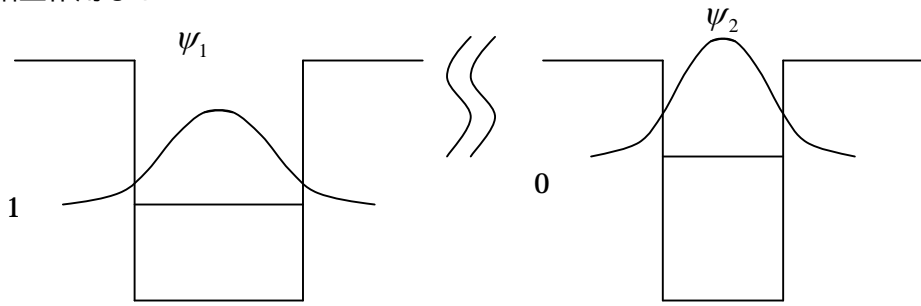
コヒーレント振動

ここで相互作用 off にすると電子が $|E_2\rangle$ に残る
エネルギー保存 (実励起)

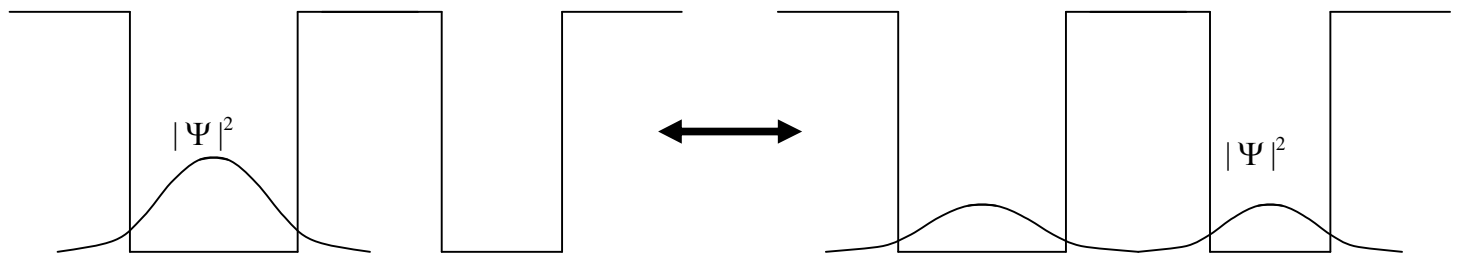
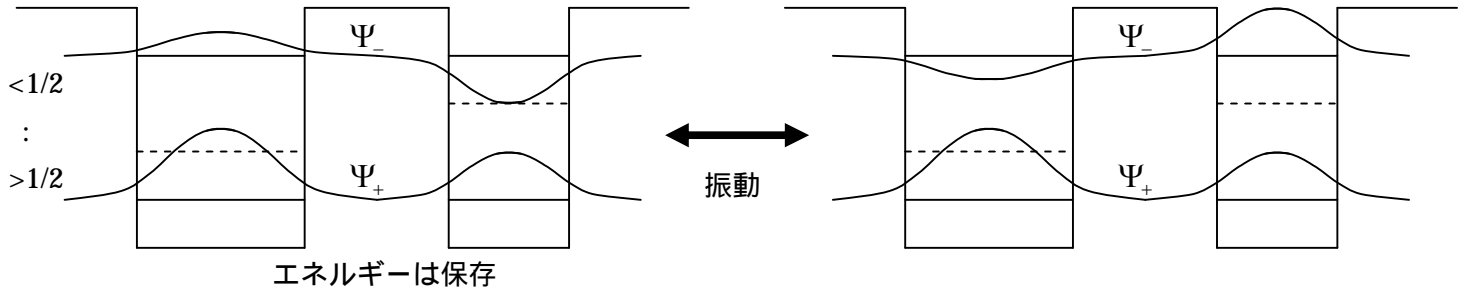


非共鳴のとき、 $\omega < \omega_0$ $E_1 + (n+1)\hbar\omega < E_2 + n\hbar\omega$

相互作用なし



相互作用あり

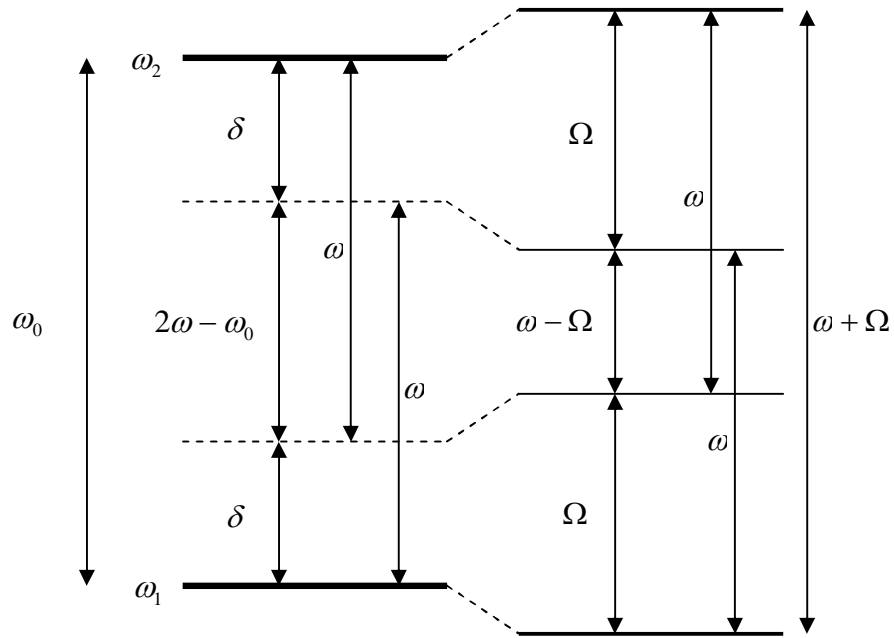
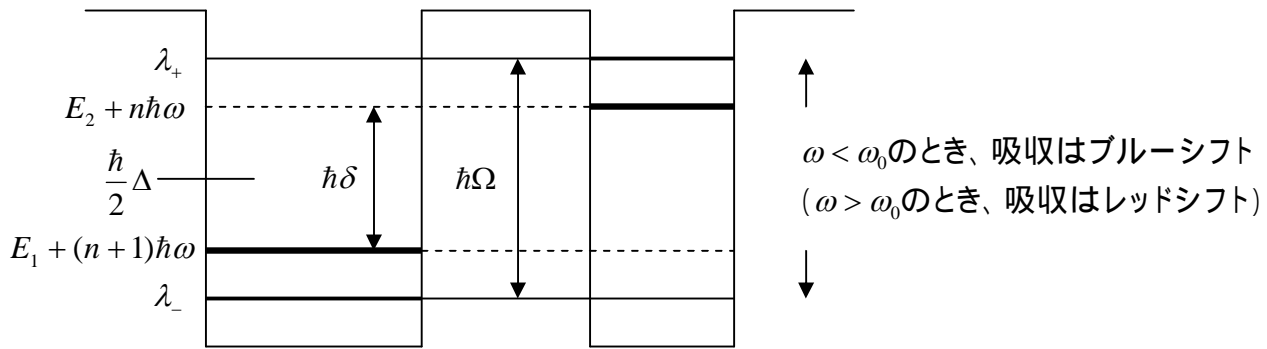


コヒーレント振動

ここで相互作用 off にするとエネルギー保存しないので電子は $|E_1\rangle$ に戻る (仮想励起)

$$\omega < \omega_0 \quad E_1 + (n+1)\hbar\omega < E_2 + n\hbar\omega \quad \delta = \omega_0 - \omega \quad \Omega = \sqrt{\delta^2 + |\xi|^2} \quad \hbar\Delta = E_1 + E_2 + (2n+1)\hbar\omega$$

$$\lambda_{\pm} = \frac{\hbar}{2}(\Delta \pm \Omega)$$



Optical Bloch 方程式

$$\frac{d\rho_{11}}{dt} = i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_1\rho_{11} + \Gamma_2\rho_{22}$$

$$\frac{d\rho_{22}}{dt} = -i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_2\rho_{22} + \Gamma_1\rho_{11}$$

$$\frac{d\rho_{12}}{dt} = i\omega_0\rho_{12} + i\eta(\rho_{22} - \rho_{11}) - \gamma_2\rho_{12}$$

$$\frac{d\rho_{21}}{dt} = -i\omega_0\rho_{21} - i\eta^*(\rho_{22} - \rho_{11}) - \gamma_2\rho_{21}$$

詳細平衡 detailed balance

$$\frac{\rho_{22}}{\rho_{11}} = \exp\left(-\frac{E_2 - E_1}{kT}\right) = e^{-\frac{\hbar\omega_0}{kT}}$$

$$\rho_{11} \xrightarrow{\Gamma_1} \rho_{22} \quad \rho_{11} \xleftarrow{\Gamma_2} \rho_{22}$$

$$\frac{d\rho_{11}}{dt} = -\Gamma_1\rho_{11} + \Gamma_2\rho_{22} = 0 \quad \frac{d\rho_{22}}{dt} = -\Gamma_2\rho_{22} + \Gamma_1\rho_{11} = 0$$

$$\frac{\rho_{22}}{\rho_{11}} = \frac{\Gamma_1}{\Gamma_2} = e^{-\frac{\hbar\omega_0}{kT}} \quad T \approx 0 \text{ では } \Gamma_1 \ll \Gamma_2$$

$$\rho_{11} + \rho_{22} = 1 \quad \rho_{11} = \rho \quad \rho_{22} = \rho e^{-\frac{\hbar\omega_0}{kT}} \quad \rho_{11} + \rho_{22} = \rho(1 + e^{-\frac{\hbar\omega_0}{kT}}) = 1 \quad \rho = \frac{1}{1 + e^{-\frac{\hbar\omega_0}{kT}}}$$

$$W \equiv \rho_{22} - \rho_{11} \quad W_{t \rightarrow \infty} = \rho e^{-\frac{\hbar\omega_0}{kT}} - \rho = \rho(e^{-\frac{\hbar\omega_0}{kT}} - 1) = \frac{e^{-\frac{\hbar\omega_0}{kT}} - 1}{e^{-\frac{\hbar\omega_0}{kT}} + 1} \rightarrow -1 (T \rightarrow 0) \quad \frac{\Gamma_1}{\Gamma_2} \rightarrow 0 (T \rightarrow 0)$$

$$W - W_\infty = \rho_{22} - \rho_{11} - \frac{e^{-\frac{\hbar\omega_0}{kT}} - 1}{e^{-\frac{\hbar\omega_0}{kT}} + 1}$$

$$(e^{-\frac{\hbar\omega_0}{kT}} + 1)(W - W_\infty) = (e^{-\frac{\hbar\omega_0}{kT}} + 1)(1 - 2\rho_{11}) - (e^{-\frac{\hbar\omega_0}{kT}} - 1) \\ = 2 - 2\rho_{11}(e^{-\frac{\hbar\omega_0}{kT}} + 1)$$

$$-2\Gamma_2\rho_{22} + 2\Gamma_1\rho_{11} = -2\Gamma_2(\rho_{22} - \frac{\Gamma_1}{\Gamma_2}\rho_{11}) = -2\Gamma_2(1 - \rho_{11} - e^{-\frac{\hbar\omega_0}{kT}}\rho_{11})$$

$$= -2\Gamma_2[1 - \rho_{11}(1 + e^{-\frac{\hbar\omega_0}{kT}})] = -\Gamma_2[2 - 2\rho_{11}(1 + e^{-\frac{\hbar\omega_0}{kT}})] = -\Gamma_2(e^{-\frac{\hbar\omega_0}{kT}} + 1)(W - W_\infty)$$

$$\frac{dW}{dt} = [-i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_2\rho_{22} + \Gamma_1\rho_{11}] - [i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_1\rho_{11} + \Gamma_2\rho_{22}]$$

$$= -i 2(\eta\rho_{21} - \eta^*\rho_{12}) - 2\Gamma_2\rho_{22} + 2\Gamma_1\rho_{11}$$

$$= -i 2(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_2(e^{-\frac{\hbar\omega_0}{kT}} + 1)(W - W_\infty)$$

$$= -i 2(\eta\rho_{21} - \eta^*\rho_{12}) - \gamma_1(W - W_\infty)$$

$$u \equiv \rho_{12} + \rho_{21}$$

$$v \equiv i(\rho_{21} - \rho_{12})$$

$$\frac{d\rho_{11}}{dt} = i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_1\rho_{11} + \Gamma_2\rho_{22}$$

$$\frac{d\rho_{22}}{dt} = -i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_2\rho_{22} + \Gamma_1\rho_{11}$$

$$\frac{d\rho_{12}}{dt} = i\omega_0\rho_{12} + i\eta(\rho_{22} - \rho_{11}) - \gamma_2\rho_{12}$$

$$\frac{d\rho_{21}}{dt} = -i\omega_0\rho_{21} - i\eta^*(\rho_{22} - \rho_{11}) - \gamma_2\rho_{21}$$

$$W \equiv \rho_{22} - \rho_{11}$$

$$W - W_\infty = \rho_{22} - \rho_{11} - \frac{e^{-\frac{\hbar\omega_0}{kT}} - 1}{e^{-\frac{\hbar\omega_0}{kT}} + 1}$$

$$\begin{aligned} \frac{dW}{dt} &= [-i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_2\rho_{22} + \Gamma_1\rho_{11}] - [i(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_1\rho_{11} + \Gamma_2\rho_{22}] \\ &= -i2(\eta\rho_{21} - \eta^*\rho_{12}) - 2\Gamma_2\rho_{22} + 2\Gamma_1\rho_{11} \\ &= -i2(\eta\rho_{21} - \eta^*\rho_{12}) - \Gamma_2(e^{-\frac{\hbar\omega_0}{kT}} + 1)(W - W_\infty) \\ &= -i2(\eta\rho_{21} - \eta^*\rho_{12}) - \gamma_1(W - W_\infty) \end{aligned}$$

$$u \equiv \rho_{12} + \rho_{21}$$

$$\begin{aligned} \frac{du}{dt} &= i\omega_0\rho_{12} + i\eta(\rho_{22} - \rho_{11}) - \gamma_2\rho_{12} - i\omega_0\rho_{21} - i\eta^*(\rho_{22} - \rho_{11}) - \gamma_2\rho_{21} \\ &= i(\eta - \eta^*)W - \gamma_2u - \omega_0v \end{aligned}$$

$$v \equiv i(\rho_{21} - \rho_{12})$$

$$\begin{aligned} \frac{dv}{dt} &= i[-i\omega_0\rho_{21} - i\eta^*(\rho_{22} - \rho_{11}) - \gamma_2\rho_{21} - i\omega_0\rho_{12} - i\eta(\rho_{22} - \rho_{11}) + \gamma_2\rho_{12}] \\ &= \omega_0\rho_{21} + \eta^*(\rho_{22} - \rho_{11}) - i\gamma_2\rho_{21} + \omega_0\rho_{12} + \eta(\rho_{22} - \rho_{11}) + i\gamma_2\rho_{12} \\ &= (\eta^* + \eta)W - \gamma_2v + \omega_0u \end{aligned}$$

緩和がなければ

$$\begin{aligned}
 |u|^2 + |v|^2 + |w|^2 &= |\rho_{12} + \rho_{21}|^2 + |\rho_{21} - \rho_{12}|^2 + |\rho_{22} - \rho_{11}|^2 \\
 &= 2|\rho_{12}|^2 + 2|\rho_{21}|^2 + |\rho_{22}|^2 + |\rho_{11}|^2 - 2\rho_{22}\rho_{11} \\
 &= 4|C_1|^2|C_2|^2 + |C_1|^4 + |C_2|^4 - 2|C_1|^2|C_2|^2 = (|C_1|^2 + |C_2|^2)^2 = 1
 \end{aligned}$$

$$u + iv = 2\rho_{12} \quad u - iv = 2\rho_{21}$$

$$\begin{aligned}
 \frac{dW}{dt} &= -i[\eta(u - iv) - \eta^*(u + iv)] - \gamma_1(W - W_\infty) \\
 &= -i[(\eta - \eta^*)u - i(\eta + \eta^*)v] - \gamma_1(W - W_\infty)
 \end{aligned}$$

$$\frac{du}{dt} = i(\eta - \eta^*)W - \gamma_2u - \omega_0v$$

$$\frac{dv}{dt} = (\eta^* + \eta)W - \gamma_2v + \omega_0u$$

$$\begin{aligned}
 \frac{d(u + iv)}{dt} &= i(\eta - \eta^*)W - \gamma_2u - \omega_0v + i(\eta + \eta^*)W - i\gamma_2v + i\omega_0u \\
 &= i2\eta W - \gamma_2(u + iv) + i\omega_0(u + iv) \\
 &= i\xi(e^{i\omega t} + e^{-i\omega t})W - \gamma_2(u + iv) + i\omega_0(u + iv)
 \end{aligned}$$

$$\begin{aligned}
 \frac{d(u - iv)}{dt} &= i(\eta - \eta^*)W - \gamma_2u - \omega_0v - i(\eta + \eta^*)W + i\gamma_2v - i\omega_0u \\
 &= -i2\eta^*W - \gamma_2(u - iv) - i\omega_0(u - iv) \\
 &= -i\xi^*(e^{i\omega t} + e^{-i\omega t})W - \gamma_2(u - iv) - i\omega_0(u - iv)
 \end{aligned}$$

$$(u + iv)e^{-i\omega t} = U + iV$$

$$(u - iv)e^{i\omega t} = U - iV$$

$$\begin{aligned}
 \frac{dW}{dt} &= -i[\eta(u - iv) - \eta^*(u + iv)] - \gamma_1(W - W_\infty) \\
 &= -i\left[\xi \frac{e^{i\omega t} + e^{-i\omega t}}{2} (U - iV)e^{-i\omega t} - \xi^* \frac{e^{-i\omega t} + e^{i\omega t}}{2} (U + iV)e^{i\omega t} \right] - \gamma_1(W - W_\infty) \\
 &= -i\left[\frac{\xi}{2} (U - iV)(1 + e^{-i2\omega t}) - \frac{\xi^*}{2} (U + iV)(1 + e^{i2\omega t}) \right] - \gamma_1(W - W_\infty)
 \end{aligned}$$

$$\begin{aligned}
 \frac{d(U + iV)}{dt} &= \frac{d(u + iv)}{dt} e^{-i\omega t} - i\omega(U + iV) \\
 &= i\xi(1 + e^{-i2\omega t})W - \gamma_2(u + iv)e^{-i\omega t} + i\omega_0(u + iv)e^{-i\omega t} - i\omega(U + iV) \\
 &= i\xi(1 + e^{-i2\omega t})W - \gamma_2(U + iV) + i(\omega_0 - \omega)(U + iV)
 \end{aligned}$$

回転波近似

$$\frac{dW}{dt} \cong -i\left[\frac{\xi}{2}(U - iV) - \frac{\xi^*}{2}(U + iV) \right] - \gamma_1(W - W_\infty)$$

$$\frac{d(U + iV)}{dt} \cong i\xi W - \gamma_2(U + iV) + i(\omega_0 - \omega)(U + iV)$$

$\frac{dW}{dt} \cong (\text{Im } \xi)U - (\text{Re } \xi)V - \gamma_1(W - W_\infty)$ $\frac{dU}{dt} \cong -(\text{Im } \xi)W - \gamma_2U - (\omega_0 - \omega)V$ $\frac{dV}{dt} \cong (\text{Re } \xi)W - \gamma_2V + (\omega_0 - \omega)U$	Bloch方程式
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Bloch方程式

$$\frac{dU}{dt} = -(\text{Im } \xi)W - (\omega_0 - \omega)V - \gamma_2 U$$

$$\frac{dV}{dt} = (\text{Re } \xi)W + (\omega_0 - \omega)U - \gamma_2 V$$

$$\frac{dW}{dt} = (\text{Im } \xi)U - (\text{Re } \xi)V - \gamma_1(W - W_\infty)$$

$$\mathbf{u} = \begin{pmatrix} U \\ V \\ W \end{pmatrix} \quad \mathbf{\Omega} = \begin{pmatrix} -\text{Re } \xi \\ -\text{Im } \xi \\ \omega_0 - \omega \end{pmatrix}$$

$$\frac{d\mathbf{u}}{dt} = \mathbf{\Omega} \times \mathbf{u} + (\text{緩和項})$$

$$\mathbf{\Omega} \times \mathbf{u} = \begin{vmatrix} \mathbf{i} & \mathbf{j} & \mathbf{k} \\ -\text{Re } \xi & -\text{Im } \xi & \omega_0 - \omega \\ U & V & W \end{vmatrix}$$

$$E_0 = |E_0| e^{i\theta}$$

$$\text{光電場 } E = |E_0| \cos(\omega t + \theta)$$

$$\xi = \frac{\mu_{12} E_0}{\hbar}$$

$$\mu_{12} = |\mu_{12}| e^{i\varphi}$$

$$\theta + \varphi = 0 \text{ のとき、 } \text{Re } \xi = \xi \quad \text{Im } \xi = 0$$

定常解

$$-(\omega_0 - \omega)V - \gamma_2 U = 0$$

$$\xi W + (\omega_0 - \omega)U - \gamma_2 V = 0$$

$$-\xi V - \gamma_1(W - W_\infty) = 0$$

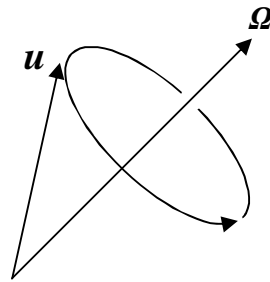
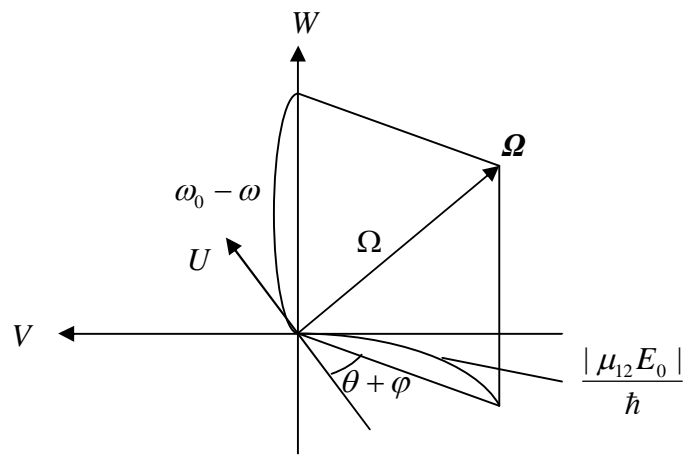
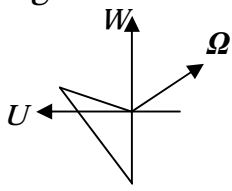
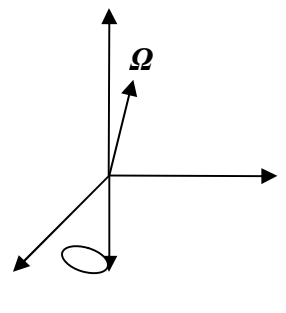
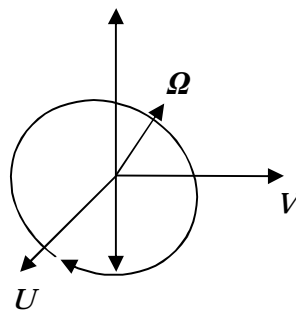
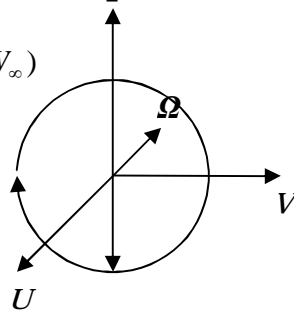
$$V = -\frac{\gamma_1}{\xi}(W - W_\infty)$$

$$U = -\frac{(\omega_0 - \omega)}{\gamma_2} V$$

$$W = \frac{(\omega_0 - \omega)^2 + \gamma_2^2}{\xi \gamma_2} V = \left[\frac{(\omega_0 - \omega)^2 + \gamma_2^2}{\xi \gamma_2} W \right] \left[-\frac{\gamma_1}{\xi}(W - W_\infty) \right]$$

$$= -\frac{\gamma_1}{\xi} \left[\frac{(\omega_0 - \omega)^2 + \gamma_2^2}{\xi \gamma_2} \right] (W - W_\infty)$$

$$W = \frac{\frac{\gamma_1}{\xi} \left[\frac{(\omega_0 - \omega)^2 + \gamma_2^2}{\xi \gamma_2} \right]}{1 + \frac{\gamma_1}{\xi} \left[\frac{(\omega_0 - \omega)^2 + \gamma_2^2}{\xi \gamma_2} \right]} W_\infty$$



緩和があるとき t が十分大きいところで Bloch ベクトルは静止したベクトルになる (回転系で静止 = 回転している)

$\omega = \omega_0$ $W_\infty = -1$ $\gamma_1 = 2\gamma_2$ $\frac{\gamma_2}{\xi} = 1$ のとき

$$W = \frac{-\frac{2\gamma_2^2}{\xi^2}}{1 + \frac{2\gamma_2^2}{\xi^2}} = -\frac{2}{3}$$

$$W = \rho_{22} - \rho_{11} = -\frac{2}{3} \quad \rho_{22} + \rho_{11} = 1$$

$$\rho_{22} = \frac{1}{6} \quad \rho_{11} = \frac{5}{6}$$

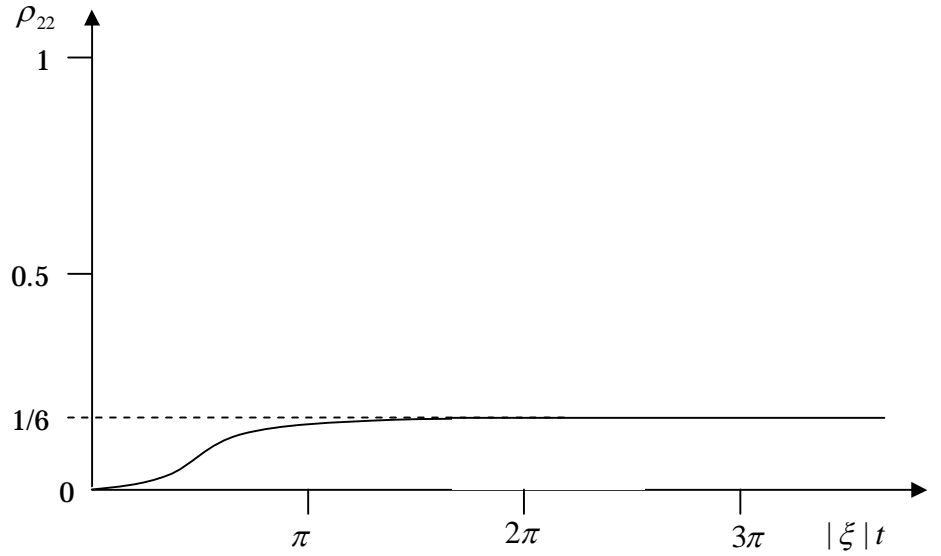
$$V = -\frac{2\gamma_2}{\xi}(W + 1) = -\frac{2}{3}$$

$$U = 0$$

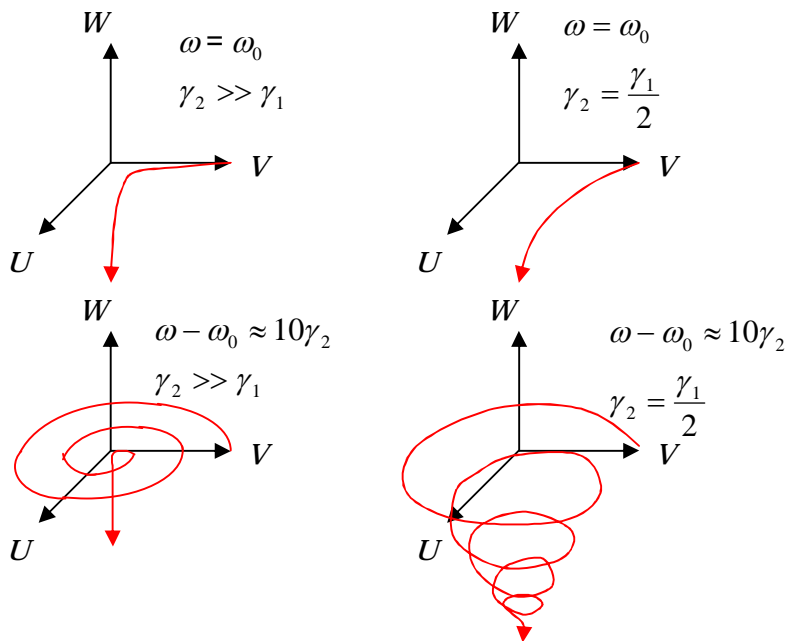
$$U + iV = (u + iv)e^{-i\omega t}$$

$$2\rho_{12} = u + iv = -\frac{2}{3}ie^{i\omega t}$$

$$\rho_{12} = -\frac{1}{3}ie^{i\omega t}$$



$t = 0$ でBloch Vector $\begin{pmatrix} U \\ V \\ W \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}$ 外場offになったときのBloch Vectorの変化



$$\gamma_2 = \left(\frac{\Gamma_1}{2} + \frac{\Gamma_2}{2}\right) + \gamma_p$$

Γ_1, Γ_2 : 準位1,2のエネルギー緩和率 (energy or population relaxation rate), 縦緩和

γ_2 : 1-2間の分極の位相緩和率 (phase relaxation rate, polarization decay rate), 横緩和

γ_p : pure dephasing rate (エネルギー緩和しないで位相のみ緩和する)

$$\begin{cases} \frac{dC_1}{dt} = -i\omega_1 C_1 + i\eta C_2 - \frac{\Gamma_1}{2} C_1 \\ \frac{dC_2}{dt} = -i\omega_2 C_2 + i\eta^* C_1 - \frac{\Gamma_2}{2} C_2 \end{cases}$$

$$\frac{d\rho_{22}}{dt} = \frac{dC_2}{dt} C_2^* + C_2 \frac{dC_2^*}{dt} = i(\eta^* \rho_{12} - \eta \rho_{21}) - \Gamma_2 \rho_{22}$$

$$\frac{d\rho_{11}}{dt} = i(\eta \rho_{21} - \eta^* \rho_{12}) - \Gamma_1 \rho_{11}$$

$$\frac{d\rho_{12}}{dt} = \frac{dC_1}{dt} C_2^* + C_1 \frac{dC_2^*}{dt} = i(\omega_2 - \omega_1) \rho_{12} + i\eta(\rho_{22} - \rho_{11}) - \left(\frac{\Gamma_1}{2} + \frac{\Gamma_2}{2}\right) \rho_{12}$$

↓

$$\gamma_2 = \frac{\Gamma_1}{2} + \frac{\Gamma_2}{2} + \gamma_p$$

準位1が基底状態 $\Gamma_1 = 0$ のとき $\gamma_2 = \frac{\Gamma_2}{2} + \gamma_p$ $\Gamma_2 = \gamma_1$ とおく $\gamma_2 = \frac{\gamma_1}{2} + \gamma_p$ $\gamma_p = 0$ のときは γ_2 は γ_1 のみで決まる

$$\text{分極 } P(t) = P_0 e^{i\omega_0 t - \gamma_2 t}$$

$$\text{population } \propto |P(t)|^2 = P_0^2 e^{-2\gamma_2 t} = P_0^2 e^{-\gamma_1 t}$$

吸収線幅がエネルギー緩和時間でなく位相緩和時間で決まる理由

$$P(\omega) = \varepsilon_0 \chi(\omega) E(\omega)$$

$$P(t) = \varepsilon_0 \int_0^\infty d\tau \chi(\tau) E(t-\tau) = \varepsilon_0 \int_{-\infty}^t d\tau \chi(t-\tau) E(\tau)$$

$t > 0$ のとき

$$\chi(\omega) \propto \frac{1}{\omega_0^2 - \omega^2 - i\omega\Gamma_0} \quad \gamma_2 = \Gamma_0 / 2$$

$$\chi(\omega) \propto \frac{\omega_0^2 - \omega^2}{(\omega_0^2 - \omega^2)^2 + \omega^2 \Gamma_0^2} + i \frac{\omega \Gamma_0}{(\omega_0^2 - \omega^2)^2 + \omega^2 \Gamma_0^2}$$

$\omega \sim \omega_0$ 付近の近似式

$$\chi(\omega) \propto \frac{(\omega_0 + \omega)(\omega_0 - \omega)}{(\omega_0 + \omega)^2 (\omega_0 - \omega)^2 + \omega^2 \Gamma_0^2} + i \frac{\omega \Gamma_0}{(\omega_0 + \omega)^2 (\omega_0 - \omega)^2 + \omega^2 \Gamma_0^2}$$

$$\sim \frac{2\omega_0(\omega_0 - \omega)}{4\omega_0^2 (\omega_0 - \omega)^2 + \omega_0^2 \Gamma_0^2} + i \frac{\omega_0 \Gamma_0}{4\omega_0^2 (\omega_0 - \omega)^2 + \omega_0^2 \Gamma_0^2}$$

$$= \frac{(\omega_0 - \omega)}{2\omega_0 (\omega_0 - \omega)^2 + 2\omega_0 (\Gamma_0 / 2)^2} + i \frac{\Gamma_0 / 2}{2\omega_0 (\omega_0 - \omega)^2 + 2\omega_0 (\Gamma_0 / 2)^2}$$

$$\propto \frac{(\omega_0 - \omega)}{(\omega_0 - \omega)^2 + (\Gamma_0 / 2)^2} + i \frac{\Gamma_0 / 2}{(\omega_0 - \omega)^2 + (\Gamma_0 / 2)^2} \quad \text{虚部が吸収スペクトルに比例 半値半幅} \Gamma_0 / 2$$

$$\chi(t) = \frac{1}{2\pi} \int \chi(\omega) e^{-i\omega t} d\omega \propto \int \frac{e^{-i\omega t}}{\omega_0^2 - \omega^2 - i\omega\Gamma_0} d\omega$$

$$\chi(t) = \frac{1}{2\pi} \int \chi(\omega) e^{-i\omega t} d\omega = \frac{1}{2\pi} \frac{1}{\varepsilon_0} \frac{N_0}{V} \frac{q^2}{m} \int \frac{e^{-i\omega t}}{\omega_0^2 - \omega^2 - i\omega\Gamma_0} d\omega = \frac{1}{\varepsilon_0} \frac{N_0}{V} \frac{q^2}{m} \frac{e^{-\frac{\Gamma_0}{2}t}}{\sqrt{\omega_0^2 - \frac{\Gamma_0^2}{4}}} \sin\left(\sqrt{\omega_0^2 - \frac{\Gamma_0^2}{4}} t\right)$$

$t < 0$ のときは 複素 ω 平面の上半平面での複素積分 より $\chi(t) = 0$

広帯域入射光 $E(t) = E_0 \delta(t)$ に対する応答 \rightarrow フーリエ変換 flatなスペクトル

$$P(t) = \varepsilon_0 \int_0^\infty d\tau \chi(\tau) E(t-\tau) = \varepsilon_0 \int_0^\infty d\tau \chi(\tau) E_0 \delta(t-\tau) = \varepsilon_0 E_0 \chi(t) \quad \text{周波数} \omega = \sqrt{\omega_0^2 - \frac{\Gamma_0^2}{4}} \text{ の分極が } e^{-\gamma_2 t} \text{ で減衰}$$

この分極による 2次波がちょうど入射光を逆位相で打ち消す \rightarrow 透過の減少 = 吸収

$\omega_0 \gg \Gamma_0 / 2$ なので $\omega \approx \omega_0$

減衰分極振動 $\chi(t) \propto \text{Re}[i\theta(t)e^{-i\omega_0 t - \gamma_2 t}]$ のフーリエ変換

$$i \int_{-\infty}^\infty \theta(t) e^{-i\omega_0 t - \gamma_2 t} e^{i\omega t} dt = i \int_0^\infty e^{-i(\omega_0 - \omega)t - \gamma_2 t} dt = i \frac{-1}{-i(\omega_0 - \omega) - \gamma_2} = \frac{1}{(\omega_0 - \omega) - i\gamma_2}$$

の虚部は $\gamma_2 (= \Gamma_0 / 2)$ の半値半幅を持つ。つまり吸収線の幅は位相緩和時間

(polarization decay time) で決まる。