

## 積層膜からの和周波発生(SFG) — 表式

### 準備

原点：光線の入射点を取る。（広がりがある場合にはその中央）

z 軸：媒質 1 から媒質 2 に向けた法線。

x 軸：界面上でみた光の進行方向にとる。（ $\mathbf{k}$  ベクトルの界面への射影に沿わせる）

次のように用語を定義する。

1/m 界面：媒質-1 と膜の界面。

m/2 界面：媒質-1 と膜の界面。

$n_1, n_2, n_m$ ：媒質-1、媒質-2、膜 m の屈折率。

$k_1, k_2, k_m$ ：媒質-1、媒質-2、膜 m の中で波動ベクトル。

$\theta_1, \theta_2, \theta_m$ ：媒質-1、媒質-2、膜 m の中を進む光の光路が法線に対してなす角。

$t_{1m}$ ：媒質-1 側から膜側に透過する光の電場に対する透過係数。

$t_{m1}$ ：膜側から媒質-1 側に透過する光の電場に対する透過係数。

$r_{1m}$ ：媒質-1 側から来て 1/m 界面で反射する光の電場に対する反射係数。

$r_{m1}$ ：膜側から来て m/1 界面で反射する光の電場に対する反射係数。

$t_{m2}$ ：膜側から媒質-2 側に透過する光の電場に対する透過係数。

$t_{2m}$ ：媒質-2 側から膜側に透過する光の電場に対する透過係数。

$r_{m2}$ ：膜側から来て m/2 界面で反射する光の電場に対する反射係数。

$r_{2m}$ ：媒質-2 側から来て 1/m 界面で反射する光の電場に対する反射係数。

上向き(-)光：m/2 界面から m/1 界面に向かう光。反射光と同じく、-z 側に進む。

下向き(+)光：m/1 界面から m/2 界面に向かう光。入射光と同じく、+z 側に進む。

### （反射係数）

$$r_{12}^s = E_r^s/E_i^s = (k_{1z} - k_{2z})/(k_{1z} + k_{2z}) = (n_1 \cos \theta_1 - n_2 \cos \theta_2)/(n_1 \cos \theta_1 + n_2 \cos \theta_2)$$

$$= -\sin(\theta_1 - \theta_2)/\sin(\theta_1 + \theta_2)$$

$$r_{12}^p = E_r^p/E_i^p = (\epsilon_1 k_{2z} - \epsilon_2 k_{1z})/(\epsilon_2 k_{1z} + \epsilon_1 k_{2z}) = (n_1 \cos \theta_2 - n_2 \cos \theta_1)/(n_2 \cos \theta_1 + n_1 \cos \theta_2)$$

$$= -\tan(\theta_1 - \theta_2)/\tan(\theta_1 + \theta_2)$$

### （透過係数）

$$t_{12}^s = E_t^s/E_i^s = 2k_{1z}/(k_{1z} + k_{2z}) = 2n_1 \cos \theta_1/(n_1 \cos \theta_1 + n_2 \cos \theta_2)$$

$$= 2 \cos \theta_1 \sin \theta_2 / \sin(\theta_1 + \theta_2)$$

$$t_{12}^p = E_t^p/E_i^p = 2\epsilon_1 k_{2z} \cos \theta_1 / (\epsilon_2 k_{1z} + \epsilon_1 k_{2z}) = 2n_1 \cos \theta_1 / (n_2 \cos \theta_1 + n_1 \cos \theta_2)$$

$$= 2 \cos \theta_1 \sin \theta_2 / \sin(\theta_1 + \theta_2) \cos(\theta_1 - \theta_2)$$

$$r_{21}^s = -r_{12}^s, \quad r_{21}^p = -r_{12}^p$$

$$t_{21}^s = (n_2 \cos \theta_2 / n_1 \cos \theta_1) t_{12}^s, \quad t_{21}^p = (n_2 \cos \theta_2 / n_1 \cos \theta_1) t_{12}^p$$

界面の内向き法線を z 軸に、s 偏光の電場の方向を y 軸に、光の進行方向が正になるように x 軸を定義して、電場の軸方向成分に対する係数を求めると、次のようになる。

$$\begin{aligned}
E_{x,i} &= E_i^p \cos \theta_1, & E_{y,i} &= E_i^s, & E_{z,i} &= -E_i^p \sin \theta_1 \\
E_{x,r} &= E_r^p \cos \theta_1, & E_{y,r} &= E_r^s, & E_{z,r} &= E_r^p \sin \theta_1 \\
E_{x,t} &= E_t^p \cos \theta_2, & E_{y,t} &= E_t^s, & E_{z,t} &= -E_t^p \sin \theta_2
\end{aligned}$$

上の表式より、下の振幅反射係数および振幅等価係数の表式を得る。

$$\begin{aligned}
r_{12}^x &= r_{12}^p, & r_{12}^y &= r_{12}^s, & r_{12}^z &= -r_{12}^p \\
r_{21}^x &= -r_{12}^p, & r_{21}^y &= -r_{12}^s, & r_{21}^z &= r_{12}^p \\
t_{12}^x &= (\cos \theta_2 / \cos \theta_1) t_{12}^p, & t_{12}^y &= t_{12}^s, & t_{12}^z &= (\sin \theta_2 / \sin \theta_1) t_{12}^p = (n_1 / n_2) t_{12}^p \\
t_{21}^x &= (n_2 / n_1) t_{12}^p, & t_{21}^y &= t_{12}^s, & t_{21}^z &= (\cos \theta_2 / \cos \theta_1) (n_2 / n_1)^2 t_{12}^p \\
r_{1m,x} &= -r_{m1,x} = r_{1m,p}, & r_{1m,y} &= -r_{m1,y} = r_{1m,s}, & r_{1m,z} &= -r_{m1,z} = -r_{1m,p} \\
r_{2m,x} &= -r_{m2,x} = r_{2m,p}, & r_{2m,y} &= -r_{m2,y} = r_{2m,s}, & r_{2m,z} &= -r_{m2,z} = -r_{2m,p} \\
t_{1m,x} &= (\cos \theta_m / \cos \theta_1) t_{1m,p}, & t_{1m,y} &= t_{1m,s}, & t_{1m,z} &= (\sin \theta_m / \sin \theta_1) t_{1m,p} \\
t_{m1,x} &= (\cos \theta_1 / \cos \theta_m) t_{m1,p}, & t_{m1,y} &= t_{m1,s}, & t_{m1,z} &= (\sin \theta_1 / \sin \theta_m) t_{m1,p} \\
t_{2m,x} &= (\cos \theta_m / \cos \theta_2) t_{2m,p}, & t_{2m,y} &= t_{2m,s}, & t_{2m,z} &= (\sin \theta_m / \sin \theta_2) t_{2m,p} \\
t_{m2,x} &= (\cos \theta_2 / \cos \theta_m) t_{m2,p}, & t_{m2,y} &= t_{m2,s}, & t_{m2,z} &= (\sin \theta_2 / \sin \theta_m) t_{m2,p} \\
t_{1m,\alpha} t_{m1,\alpha} &= 1 + r_{1m,\alpha} r_{m1,\alpha} = 1 - r_{1m,\alpha}^2, & t_{2m,\alpha} t_{m2,\alpha} &= 1 + r_{2m,\alpha} r_{m2,\alpha} = 1 - r_{2m,\alpha}^2, & (\alpha = x, y, z)
\end{aligned}$$

上式は次のような関係式を満足する。

$$\begin{aligned}
1 + r_{12}^x &= t_{12}^x, & 1 + r_{12}^y &= t_{12}^y, & 1 + r_{12}^z &= (n_2 / n_1)^2 t_{12}^z \quad (\text{電場に対する境界条件}) \\
1 + r_{12}^s r_{21}^s &= 1 - (r_{12}^s)^2 = t_{12}^s t_{21}^s, & 1 + r_{12}^p r_{21}^p &= 1 - (r_{12}^p)^2 = t_{12}^p t_{21}^p
\end{aligned}$$

因みに、光の強度は  $E^2 n \cos \theta$  に比例するので、 $r_{12}^2 n_1 \cos \theta_1 + t_{12}^2 n_2 \cos \theta_2 = n_1 \cos \theta_1$  となり、光の強度は保存される。

## L係数、電場振幅

$$E_{\text{SF},\alpha} = \sum_{\beta} L_{\alpha\beta} P_{\beta}^{\text{SF}}$$

共通因子である  $4\pi i \omega_{\text{SF}} / c$  (屈折率の代わりに波数ベクトルを使うときには  $4\pi i \omega_{\text{SF}}^2 / c^2$ ) を省略する。

**L係数の表記法** ;  $L_{ij,s(p),\alpha}$  : 分極シート  $m'$  が  $i$  層と  $j$  層に挟まれているときに、分極の  $\alpha$  成分 ( $\alpha = x, y, z$ ) が作る光の  $s$  偏光成分又は  $p$  偏光成分の間の係数 ( $x, y, z$  成分ではないことに注意)。上向き (-) 光、上向き (+) 光の区別を上付き -, + で示す。

**媒質 1 と積層膜  $m$  の間の分極シート  $m'$  からの光生成** に対しては、

$$\begin{aligned}
L_{1/m,p,x}^- &= \cos \theta_{m,\text{SF}} / (n_{1,\text{SF}} \cos \theta_{m,\text{SF}} + n_{m,\text{SF}} \cos \theta_{1,\text{SF}}) \\
L_{1/m,s,y}^- &= 1 / (n_{1,\text{SF}} \cos \theta_{1,\text{SF}} + n_{m,\text{SF}} \cos \theta_{m,\text{SF}}) \\
L_{1/m,p,z}^- &= (n_{m,\text{SF}} / n_{m',\text{SF}}) \sin \theta_{m',\text{SF}} / (n_{1,\text{SF}} \cos \theta_{m,\text{SF}} + n_{m,\text{SF}} \cos \theta_{1,\text{SF}})
\end{aligned}$$

$$\begin{aligned}
&= (n_{m,SF}/n_{m',SF})^2 \sin\theta_{m,SF}/(n_{1,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{1,SF}) \\
L_{1/m,p,x}^+ &= \cos\theta_{1,SF}/(n_{1,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{1,SF}) \\
L_{1/m,s,y}^+ &= 1/(n_{1,SF}\cos\theta_{1,SF} + n_{m,SF}\cos\theta_{m,SF}) \\
L_{1/m,p,z}^+ &= -(n_{1,SF}/n_{m',SF})\sin\theta_{m',SF}/(n_{1,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{1,SF}) \\
&= -(n_{1,SF}/n_{m',SF})^2 \sin\theta_{1,SF}/(n_{1,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{1,SF})
\end{aligned}$$

媒質 2 と積層膜 m の間の分極シート m' から光生成に対しては、

$$\begin{aligned}
L_{2/m,p,x}^- &= \cos\theta_{2,SF}/(n_{2,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{2,SF}) \\
L_{2/m,s,y}^- &= 1/(n_{2,SF}\cos\theta_{2,SF} + n_{m,SF}\cos\theta_{m,SF}) \\
L_{2/m,p,z}^- &= (n_{2,SF}/n_{m'',SF})\sin\theta_{m'',SF}/(n_{2,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{2,SF}) \\
&= (n_{2,SF}/n_{m'',SF})^2 \sin\theta_{2,SF}/(n_{2,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{2,SF}) \\
L_{2/m,p,x}^+ &= \cos\theta_{m,SF}/(n_{2,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{2,SF}) \\
L_{2/m,s,y}^+ &= 1/(n_{2,SF}\cos\theta_{2,SF} + n_{m,SF}\cos\theta_{m,SF}) \\
L_{2/m,p,z}^+ &= -(n_{m,SF}/n_{m'',SF})\sin\theta_{m'',SF}/(n_{2,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{2,SF}) \\
&= -(n_{m,SF}/n_{m'',SF})^2 \sin\theta_{m,SF}/(n_{2,SF}\cos\theta_{m,SF} + n_{m,SF}\cos\theta_{2,SF})
\end{aligned}$$

積層膜内部の分極シートからの光生成に対しては、

$$\begin{aligned}
L_{m/m,p,x}^- &= L_{m/m,p,x}^+ = \cos\theta_{m,SF}/(2n_{m,SF}\cos\theta_{m,SF}) \\
L_{m/m,s,y}^- &= L_{m/m,s,y}^+ = 1/(2n_{m,SF}\cos\theta_{m,SF}) \\
L_{m/m,p,z}^- &= -L_{m/m,p,z}^+ = \sin\theta_{m,SF}/(2n_{m,SF}\cos\theta_{m,SF})
\end{aligned}$$

よって、各部位からの SFG 光の電場振幅は次のように表される。

**$P(0)$  に由来するもの：** ( $P(0^-)$  は媒質 1 側の電場による分極、 $P(0^+)$  は膜側の電場による分極)

$$\begin{aligned}
E_{1,p}(z=0) &= L_{1/m,p,x}^- P_x(0^-) + L_{1/m,p,z}^- P_z(0^-) \\
E_{1,s}(z=0) &= L_{1/m,s,y}^- P_y(0^-) \\
E_{m,p}^+(z=0^+) &= L_{1/m,p,x}^+ P_x(0^+) + L_{1/m,p,z}^+ P_z(0^+) \\
E_{m,s}^+(z=0^+) &= L_{1/m,s,y}^+ P_y(00^+)
\end{aligned}$$

**$P(h_m)$  に由来するもの：** ( $P(h_m^+)$  は媒質 2 側の電場による分極、 $P(h_m^-)$  は膜側の電場による分極)

$$\begin{aligned}
E_{m,p}^-(z=h_m^-) &= L_{2/m,p,x}^- P_x(h_m^-) + L_{2/m,p,z}^- P_z(h_m^-) \\
E_{m,s}^-(z=h_m^-) &= L_{2/m,s,y}^- P_y(h_m^-) \\
E_{2,p}^+(z=h_m^+) &= L_{2/m,p,x}^+ P_x(h_m^+) + L_{2/m,p,z}^+ P_z(h_m^+) \\
E_{2,s}^+(z=h_m^+) &= L_{2/m,s,y}^+ P_y(h_m^+)
\end{aligned}$$

**$P(z)$  に由来するもの：**

$$\begin{aligned}
E_{m,p}(z) &= L_{m/m,p,x}^- P_x(z) + L_{m/m,p,z}^- P_z(z) \\
E_{m,s}(z) &= L_{m/m,s,y}^- P_y(z)
\end{aligned}$$

$$E_{m,p}^+(z) = L_{m,m,p,x}^+ P_x(z) + L_{m,m,p,z}^+ P_z(z)$$

$$E_{m,s}^+(z) = L_{m,m,s,y}^+ P_y(z)$$

1/m 界面の外側 ( $x = -2nh_m \tan \theta_{m,SF}, z = 0$ ) からの SFG

反射 SFG

$$E_{1p}^-(0^-) = \sum_{\beta\gamma} E_{vis,\beta}^0 E_{ir,\gamma}^0 \frac{(1+r_{1m,vis,\beta})(1+r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma})(1+r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{(1+r_{1m,vis,\beta} r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})} \\ \times \frac{(1+r_{m2,SF,p} e^{2i\beta_{m,SF} h_m}) L_{1/m,p,x}^- \chi_{x\beta\gamma} + (1-r_{m2,SF,p} e^{2i\beta_{m,SF} h_m}) L_{1/m,p,z}^- \chi_{z\beta\gamma}}{(1+r_{1m,SF,p} r_{m2,SF,p} e^{2i\beta_{m,SF} h_m})} \quad (1a)$$

$$E_{1s}^-(0^-) = \sum_{\beta\gamma} E_{vis,\beta}^0 E_{ir,\gamma}^0 \frac{(1+r_{1m,vis,\beta})(1+r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma})(1+r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{(1+r_{1m,vis,\beta} r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})} \\ \times \frac{(1+r_{m2,SF,s} e^{2i\beta_{m,SF} h_m})}{(1+r_{1m,SF,s} r_{m2,SF,s} e^{2i\beta_{m,SF} h_m})} L_{1/m,s,y}^- \chi_{y\beta\gamma} \quad (1b)$$

透過 SFG

$$E_{2p}^+(h_m^+) = \sum_{\beta\gamma} E_{vis,\beta}^0 E_{ir,\gamma}^0 \frac{(1+r_{1m,vis,\beta})(1+r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma})(1+r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{(1+r_{1m,vis,\beta} r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})} \\ \times \frac{t_{1m,SF,p} t_{m2,SF,p} e^{i\beta_{m,SF} h_m}}{1+r_{1m,SF,p} r_{m2,SF,p} e^{2i\beta_{m,SF} h_m}} [\chi_{x\beta\gamma} L_{m/1,p,x}^+ + \chi_{z\beta\gamma} L_{m/1,p,z}^+] \quad (2a)$$

$$E_{2s}^+(h_m^+) = \sum_{\beta\gamma} E_{vis,\beta}^0 E_{ir,\gamma}^0 \frac{(1+r_{1m,vis,\beta})(1+r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma})(1+r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{(1+r_{1m,vis,\beta} r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})} \\ \times \frac{t_{1m,SF,s} t_{m2,SF,s} e^{i\beta_{m,SF} h_m}}{1+r_{1m,SF,s} r_{m2,SF,s} e^{2i\beta_{m,SF} h_m}} \chi_{y\beta\gamma} L_{m/1,s,y}^+ \quad (2b)$$

表面が薄膜として完成しているときの薄膜（屈折率  $m'$ ）を形成する分子からの SFG は、内部反射を考慮する必要がある。

反射 SFG

$$E_{1p}^-(0^-) = \sum_{\beta\gamma} E_{vis,\beta}^0 E_{ir,\gamma}^0 \frac{t_{1m',vis,\beta} (1+r_{m'm,vis,\beta})(1+r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})}{1+r_{1m',vis,\beta} r_{m'm,vis,\beta} + (r_{m'm,vis,\beta} + r_{1m',vis,\beta}) r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m}} \\ \times \frac{t_{1m',ir,\gamma} (1+r_{m'm,ir,\gamma})(1+r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{1+r_{1m',ir,\gamma} r_{m'm,ir,\gamma} + (r_{m'm,ir,\gamma} + r_{1m',ir,\gamma}) r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m}} \\ \times \frac{(1+r_{1m',SF,p} r_{m'm,SF,p})(1+r_{m2,SF,p} e^{2i\beta_{m,SF} h_m}) \chi_{x\beta\gamma} L_{1/m,p,x}^- + (1-r_{m2,SF,p} e^{2i\beta_{m,SF} h_m}) \chi_{z\beta\gamma} L_{1/m,p,z}^-}{1+r_{1m',SF,p} r_{m'm,SF,p} + r_{m2,SF,p} (r_{1m',SF,p} + r_{m'm,SF,p}) e^{2i\beta_{m,SF} h_m}} \quad (3a)$$

$$\begin{aligned}
E_{1s}^{-}(0^{-}) &= \sum_{\beta\gamma} E_{\text{vis},\beta}^0 E_{ir,\gamma}^0 \frac{t_{1m',\text{vis},\beta} (1 + r_{m'm,\text{vis},\beta}) (1 + r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m})}{1 + r_{1m',\text{vis},\beta} r_{m'm,\text{vis},\beta} + (r_{m'm,\text{vis},\beta} + r_{1m',\text{vis},\beta}) r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m}} \\
&\times \frac{t_{1m',ir,\gamma} (1 + r_{m'm,ir,\gamma}) (1 + r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{1 + r_{1m',ir,\gamma} r_{m'm,ir,\gamma} + (r_{m'm,ir,\gamma} + r_{1m',ir,\gamma}) r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m}} \\
&\times \frac{(1 + r_{1m'SF,s} r_{m'm'SF,s}) (1 + r_{m2SF,s} e^{2i\beta_{m,SF} h_m})}{1 + r_{1m'SF,s} r_{m'm'SF,s} + r_{m2SF,s} (r_{1m'SF,s} + r_{m'm'SF,s})} \chi_{y\beta\gamma} L_{1/m,SY}^{-}
\end{aligned} \tag{3b}$$

### 透過 SFG

$$\begin{aligned}
E_{2p}^{+}(h_m^{+}) &= \sum_{\beta\gamma} E_{\text{vis},\beta}^0 E_{ir,\gamma}^0 \frac{t_{1m',\text{vis},\beta} (1 + r_{m'm,\text{vis},\beta}) (1 + r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m})}{1 + r_{1m',\text{vis},\beta} r_{m'm,\text{vis},\beta} + (r_{m'm,\text{vis},\beta} + r_{1m',\text{vis},\beta}) r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m}} \\
&\times \frac{t_{1m',ir,\gamma} (1 + r_{m'm,ir,\gamma}) (1 + r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{1 + r_{1m',ir,\gamma} r_{m'm,ir,\gamma} + (r_{m'm,ir,\gamma} + r_{1m',ir,\gamma}) r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m}} \\
&\times \frac{t_{m2SF,p} (1 + r_{1m'SF,p} r_{m'm'SF,p}) e^{i\beta_{m,SF} h_m}}{1 + r_{1m'SF,p} r_{m'm'SF,p} + r_{m2SF,p} (r_{mm'SF,p} + r_{m1'SF,p})} (L_{1/m,px}^{+} \chi_{x\beta\gamma} + L_{1/m,pz}^{+} \chi_{z\beta\gamma})
\end{aligned} \tag{4a}$$

$$\begin{aligned}
E_{2s}^{+}(h_m^{+}) &= \sum_{\beta\gamma} E_{\text{vis},\beta}^0 E_{ir,\gamma}^0 \frac{t_{1m',\text{vis},\beta} (1 + r_{m'm,\text{vis},\beta}) (1 + r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m})}{1 + r_{1m',\text{vis},\beta} r_{m'm,\text{vis},\beta} + (r_{m'm,\text{vis},\beta} + r_{1m',\text{vis},\beta}) r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m}} \\
&\times \frac{t_{1m',ir,\gamma} (1 + r_{m'm,ir,\gamma}) (1 + r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{1 + r_{1m',ir,\gamma} r_{m'm,ir,\gamma} + (r_{m'm,ir,\gamma} + r_{1m',ir,\gamma}) r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m}} \\
&\times \frac{t_{m2SF,s} (1 + r_{1m'SF,s} r_{m'm'SF,s}) e^{i\beta_{m,SF} h_m}}{1 + r_{1m'SF,s} r_{m'm'SF,s} + r_{m2SF,s} (r_{mm'SF,s} + r_{m1'SF,s})} L_{1/m,SY}^{+} \chi_{y\beta\gamma}
\end{aligned} \tag{4b}$$

### 1/m 界面の内側 ( $x = -2nh_m \tan\theta_{m,SF}$ , $z = 0^{+}$ ) からの SFG

### 反射 SFG

$$\begin{aligned}
E_{1p}^{-}(0^{-}) &= \sum_{\beta\gamma} E_{\text{vis},\beta}^0 E_{ir,\gamma}^0 \frac{t_{1m,\text{vis},\beta} t_{1m,ir,\gamma} (1 + r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m}) (1 + r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{(1 + r_{1m,\text{vis},\beta} r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m}) (1 + r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})} \\
&\times \frac{[(1 + r_{m2SF,p} e^{2i\beta_{m,SF} h_m}) L_{1/m,px}^{-} \chi_{x\beta\gamma} + (1 - r_{m2SF,p} e^{2i\beta_{m,SF} h_m}) L_{1/m,pz}^{-} \chi_{z\beta\gamma}]}{1 + r_{1m,SF,p} r_{m2SF,p} e^{2i\beta_{m,SF} h_m}}
\end{aligned} \tag{5a}$$

$$\begin{aligned}
E_{1s}^{-}(0^{-}) &= \sum_{\beta\gamma} E_{\text{vis},\beta}^0 E_{ir,\gamma}^0 \frac{t_{1m,\text{vis},\beta} t_{1m,ir,\gamma} (1 + r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m}) (1 + r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{(1 + r_{1m,\text{vis},\beta} r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m}) (1 + r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})} \\
&\times \frac{(1 + r_{m2SF,s} e^{2i\beta_{m,SF} h_m}) L_{1/m,SY}^{-} \chi_{y\beta\gamma}}{1 + r_{1m,SF,s} r_{m2SF,s} e^{2i\beta_{m,SF} h_m}}
\end{aligned} \tag{5b}$$

### 透過 SFG

$$\begin{aligned}
E_{2p}^{+}(h_m^{+}) &= \sum_{\beta\gamma} E_{\text{vis},\beta}^0 E_{ir,\gamma}^0 \frac{t_{1m,\text{vis},\beta} t_{1m,ir,\gamma} (1 + r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m}) (1 + r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})}{(1 + r_{1m,\text{vis},\beta} r_{m2,\text{vis},\beta} e^{2i\beta_{m,\text{vis}} h_m}) (1 + r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})} \\
&\times \frac{t_{m2SF,p} e^{i\beta_{m,SF} h_m}}{1 + r_{1m,SF,p} r_{m2SF,p} e^{2i\beta_{m,SF} h_m}} (L_{1/m,px}^{+} \chi_{x\beta\gamma} + L_{1/m,pz}^{+} \chi_{z\beta\gamma})
\end{aligned} \tag{6a}$$

$$\begin{aligned}
E^+_{2s}(h_m^+) &= \sum_{\beta, \gamma} E^0_{vis, \beta} E^0_{ir, \gamma} \frac{t_{1m, vis, \beta} t_{1m, ir, \gamma} (1 + r_{m2, vis, \beta} e^{2i\beta_{m, vis} h_m}) (1 + r_{m2, ir, \gamma} e^{2i\beta_{m, ir} h_m})}{(1 + r_{1m, vis, \beta} r_{m2, vis, \beta} e^{2i\beta_{m, vis} h_m}) (1 + r_{1m, ir, \gamma} r_{m2, ir, \gamma} e^{2i\beta_{m, ir} h_m})} \\
&\times \frac{t_{m2, SF, s} e^{i\beta_{m, SF} h_m}}{1 + r_{1m, SF, s} r_{m2, SF, s} e^{2i\beta_{m, SF} h_m}} L^{+1/m, sy} \chi_{y\beta\gamma}
\end{aligned} \tag{6b}$$

**m/2 界面の外側** ( $x = -(2n + 1)h_m \tan \theta_{m, SF}$ ,  $z = h_m^+$ ) **からの SFG**

**反射 SFG**

$$\begin{aligned}
E^-_{1p}(0^-) &= \sum_{\beta, \gamma} E^0_{vis, \beta} E^0_{ir, \gamma} \frac{t_{1m, vis, \beta} t_{m2, vis, \beta} t_{1m, ir, \gamma} t_{m2, ir, \gamma} e^{i(\beta_{m, ir} + \beta_{m, vis}) h_m}}{(1 + r_{1m, vis, \beta} r_{m2, vis, \beta} e^{2i\beta_{m, vis} h_m}) (1 + r_{1m, ir, \gamma} r_{m2, ir, \gamma} e^{2i\beta_{m, ir} h_m})} \\
&\times \frac{t_{m1, p} e^{i\beta_{m, SF} h_m}}{1 + r_{1m, p} r_{m2, p} 2e^{2i\beta_{m, SF} h_m}} [L^{-2/m, px} \chi_{x\beta\gamma} + L^{-2/m, pz} \chi_{z\beta\gamma}]
\end{aligned} \tag{7a}$$

$$\begin{aligned}
E^-_{1s}(0^-) &= \sum_{\beta, \gamma} E^0_{vis, \beta} E^0_{ir, \gamma} \frac{t_{1m, vis, \beta} t_{m2, vis, \beta} t_{1m, ir, \gamma} t_{m2, ir, \gamma} e^{i(\beta_{m, ir} + \beta_{m, vis}) h_m}}{(1 + r_{1m, vis, \beta} r_{m2, vis, \beta} e^{2i\beta_{m, vis} h_m}) (1 + r_{1m, ir, \gamma} r_{m2, ir, \gamma} e^{2i\beta_{m, ir} h_m})} \\
&\times \frac{t_{m1, s} e^{i\beta_{m, SF} h_m}}{1 + r_{1m, s} r_{m2, s} 2e^{2i\beta_{m, SF} h_m}} L^{-2/m, sy} \chi_{y\beta\gamma}
\end{aligned} \tag{7b}$$

**透過 SFG**

$$\begin{aligned}
E^+_{2p}(h_m^+) &= \sum_{\beta, \gamma} E^0_{vis, \beta} E^0_{ir, \gamma} \frac{t_{1m, vis, \beta} t_{m2, vis, \beta} t_{1m, ir, \gamma} t_{m2, ir, \gamma} e^{i(\beta_{m, ir} + \beta_{m, vis}) h_m}}{(1 + r_{1m, vis, \beta} r_{m2, vis, \beta} e^{2i\beta_{m, vis} h_m}) (1 + r_{1m, ir, \gamma} r_{m2, ir, \gamma} e^{2i\beta_{m, ir} h_m})} \\
&\times \frac{(1 - r_{1m, SF, p} e^{2i\beta_{m, SF} h_m}) L^{+2/m, px} \chi_{x\beta\gamma} + (1 + r_{1m, SF, p} e^{2i\beta_{m, SF} h_m}) L^{+2/m, pz} \chi_{z\beta\gamma}}{1 + r_{1m, SF, p} r_{m2, SF, p} e^{2i\beta_{m, SF} h_m}}
\end{aligned} \tag{8a}$$

$$\begin{aligned}
E^+_{2s}(h_m^+) &= \sum_{\beta, \gamma} E^0_{vis, \beta} E^0_{ir, \gamma} \frac{t_{1m, vis, \beta} t_{m2, vis, \beta} t_{1m, ir, \gamma} t_{m2, ir, \gamma} e^{i(\beta_{m, ir} + \beta_{m, vis}) h_m}}{(1 + r_{1m, vis, \beta} r_{m2, vis, \beta} e^{2i\beta_{m, vis} h_m}) (1 + r_{1m, ir, \gamma} r_{m2, ir, \gamma} e^{2i\beta_{m, ir} h_m})} \\
&\times \frac{(1 - r_{1m, SF, s} e^{2i\beta_{m, SF} h_m}) L^{+2/m, sy} \chi_{y\beta\gamma}}{1 + r_{1m, SF, s} r_{m2, SF, s} e^{2i\beta_{m, SF} h_m}}
\end{aligned} \tag{8b}$$

**SFG を出す界面が屈折率 m' の薄膜の時には、**

**反射 SFG**

$$\begin{aligned}
E^-_{1p}(0^-) &= \sum_{\beta, \gamma} E^0_{vis, \beta} E^0_{ir, \gamma} \frac{t_{1m, vis, \beta} t_{mm', vis, \beta} (1 + r_{m'^2, vis, \beta}) e^{i\beta_{m, vis} h_m}}{(1 + r_{mm', vis, \beta} r_{m'^2, vis, \beta}) + r_{1m, vis, \beta} (r_{mm', vis, \beta} + r_{m'^2, vis, \beta}) e^{2i\beta_{m, vis} h_m}} \\
&\times \frac{t_{1m, ir, \gamma} t_{mm', ir, \gamma} (1 + r_{m'^2, ir, \gamma}) e^{i\beta_{m, ir} h_m}}{(1 + r_{mm', ir, \gamma} r_{m'^2, ir, \gamma}) + r_{1m, ir, \gamma} (r_{mm', ir, \gamma} + r_{m'^2, ir, \gamma}) e^{2i\beta_{m, ir} h_m}} \\
&\times \frac{t_{m1, SF, p} e^{i\beta_{m, SF} h_m} (1 + r_{mm', SF, p} r_{m'^2, SF, p})}{1 + r_{mm', SF, p} r_{m'^2, SF, p} + r_{1m, SF, p} (r_{mm', SF, p} + r_{m'^2, SF, p}) e^{2i\beta_{m, SF} h_m}} [L^{-2/m, px} \chi_{x\beta\gamma} + L^{-2/m, pz} \chi_{z\beta\gamma}]
\end{aligned} \tag{9a}$$

$$\begin{aligned}
E_{1s}^{-}(0^{-}) &= \sum_{\beta\gamma} E_{\text{vis}\beta}^0 E_{\text{ir}\gamma}^0 \frac{t_{1m,\text{vis}\beta} t_{mm',\text{vis}\beta} (1+r_{m'2,\text{vis}\beta}) e^{i\beta_{m,\text{vis}} h_m}}{(1+r_{mm',\text{vis}\beta} r_{m'2,\text{vis}\beta}) + r_{1m,\text{vis}\beta} (r_{mm',\text{vis}\beta} + r_{m'2,\text{vis}\beta})} e^{2i\beta_{m,\text{vis}} h_m} \\
&\times \frac{t_{1m,\text{ir}\gamma} t_{mm',\text{ir}\gamma} (1+r_{m'2,\text{ir}\gamma}) e^{i\beta_{m,\text{ir}} h_m}}{(1+r_{mm',\text{ir}\gamma} r_{m'2,\text{ir}\gamma}) + r_{1m,\text{ir}\gamma} (r_{mm',\text{ir}\gamma} + r_{m'2,\text{ir}\gamma})} e^{2i\beta_{m,\text{ir}} h_m} \\
&\times \frac{t_{m1,\text{SF}s} e^{i\beta_{m,\text{SF}s} h_m} (1+r_{mm',\text{SF}s} r_{m'2,\text{SF}s})}{1+r_{mm',\text{SF}s} r_{m'2,\text{SF}s} + r_{1m,\text{SF}s} (r_{mm',\text{SF}s} + r_{m'2,\text{SF}s})} e^{2i\beta_{m,\text{SF}s} h_m} L^{-2/m,\text{sy}} \chi_{\beta\gamma}
\end{aligned} \tag{9b}$$

### 透過 SFG

$$\begin{aligned}
E_{2p}(h_m^{+}) &= \sum_{\beta\gamma} E_{\text{vis}\beta}^0 E_{\text{ir}\gamma}^0 \frac{t_{1m,\text{vis}\beta} t_{mm',\text{vis}\beta} (1+r_{m'2,\text{vis}\beta}) e^{i\beta_{m,\text{vis}} h_m}}{(1+r_{mm',\text{vis}\beta} r_{m'2,\text{vis}\beta}) + r_{1m,\text{vis}\beta} (r_{mm',\text{vis}\beta} + r_{m'2,\text{vis}\beta})} e^{2i\beta_{m,\text{vis}} h_m} \\
&\times \frac{t_{1m,\text{ir}\gamma} t_{mm',\text{ir}\gamma} (1+r_{m'2,\text{ir}\gamma}) e^{i\beta_{m,\text{ir}} h_m}}{(1+r_{mm',\text{ir}\gamma} r_{m'2,\text{ir}\gamma}) + r_{1m,\text{ir}\gamma} (r_{mm',\text{ir}\gamma} + r_{m'2,\text{ir}\gamma})} e^{2i\beta_{m,\text{ir}} h_m} \\
&\times \frac{(1+r_{mm',\text{SF}p} r_{m'2,\text{SF}p}) [(1-r_{1m,\text{SF}p}) e^{2i\beta_{m,\text{SF}p} h_m} L^{+2/m,\text{px}} \chi_{\beta\gamma} + (1+r_{1m,\text{SF}p}) e^{2i\beta_{m,\text{SF}p} h_m} L^{+2/m,\text{pz}} \chi_{\beta\gamma}]}{1+r_{mm',\text{SF}p} r_{m'2,\text{SF}p} + r_{1m,\text{SF}p} (r_{mm',\text{SF}p} + r_{m'2,\text{SF}p})} e^{2i\beta_{m,\text{SF}p} h_m}
\end{aligned} \tag{10a}$$

$$\begin{aligned}
E_{2s}(h_m^{+}) &= \sum_{\beta\gamma} E_{\text{vis}\beta}^0 E_{\text{ir}\gamma}^0 \frac{t_{1m,\text{vis}\beta} t_{mm',\text{vis}\beta} (1+r_{m'2,\text{vis}\beta}) e^{i\beta_{m,\text{vis}} h_m}}{(1+r_{mm',\text{vis}\beta} r_{m'2,\text{vis}\beta}) + r_{1m,\text{vis}\beta} (r_{mm',\text{vis}\beta} + r_{m'2,\text{vis}\beta})} e^{2i\beta_{m,\text{vis}} h_m} \\
&\times \frac{t_{1m,\text{ir}\gamma} t_{mm',\text{ir}\gamma} (1+r_{m'2,\text{ir}\gamma}) e^{i\beta_{m,\text{ir}} h_m}}{(1+r_{mm',\text{ir}\gamma} r_{m'2,\text{ir}\gamma}) + r_{1m,\text{ir}\gamma} (r_{mm',\text{ir}\gamma} + r_{m'2,\text{ir}\gamma})} e^{2i\beta_{m,\text{ir}} h_m} \\
&\times \frac{(1+r_{mm',\text{SF}s} r_{m'2,\text{SF}s}) (1-r_{1m,\text{SF}s}) e^{2i\beta_{m,\text{SF}s} h_m} L^{+2/m,\text{sy}} \chi_{\beta\gamma}}{1+r_{mm',\text{SF}s} r_{m'2,\text{SF}s} + r_{1m,\text{SF}s} (r_{mm',\text{SF}s} + r_{m'2,\text{SF}s})} e^{2i\beta_{m,\text{SF}s} h_m}
\end{aligned} \tag{10b}$$

### m/2 界面の内側 ( $x = -(2n+1)h_m \tan\theta_{\text{mSF}}$ , $z = h_m$ ) からの反射 SFG

#### 反射 SFG

$$\begin{aligned}
E_{1p}^{-}(0^{-}) &= \sum_{\beta\gamma} E_{\text{vis}\beta}^0 E_{\text{ir}\gamma}^0 \frac{t_{1m,\text{vis}\beta} t_{1m,\text{ir}\gamma} (1+r_{m2,\text{vis}\beta}) (1+r_{m2,\text{ir}\gamma}) e^{i(\beta_{m,\text{ir}} + \beta_{m,\text{vis}}) h_m}}{(1+r_{1m,\text{vis}\beta} r_{m2,\text{vis}\beta}) e^{2i\beta_{m,\text{vis}} h_m} (1+r_{1m,\text{ir}\gamma} r_{m2,\text{ir}\gamma}) e^{2i\beta_{m,\text{ir}} h_m}} \\
&\times \frac{t_{m1,\text{SF}p} e^{i\beta_{m,\text{SF}p} h_m}}{1+r_{1m,\text{SF}p} r_{m2,\text{SF}p}} e^{2i\beta_{m,\text{SF}p} h_m} (L^{-2/m,\text{px}} \chi_{x\beta\gamma} + L^{-2/m,\text{pz}} \chi_{z\beta\gamma})
\end{aligned} \tag{11a}$$

$$\begin{aligned}
E_{1s}^{-}(0^{-}) &= \sum_{\beta\gamma} E_{\text{vis}\beta}^0 E_{\text{ir}\gamma}^0 \frac{t_{1m,\text{vis}\beta} t_{1m,\text{ir}\gamma} (1+r_{m2,\text{vis}\beta}) (1+r_{m2,\text{ir}\gamma}) e^{i(\beta_{m,\text{ir}} + \beta_{m,\text{vis}}) h_m}}{(1+r_{1m,\text{vis}\beta} r_{m2,\text{vis}\beta}) e^{2i\beta_{m,\text{vis}} h_m} (1+r_{1m,\text{ir}\gamma} r_{m2,\text{ir}\gamma}) e^{2i\beta_{m,\text{ir}} h_m}} \\
&\times \frac{t_{m1,\text{SF}s} e^{i\beta_{m,\text{SF}s} h_m}}{1+r_{1m,\text{SF}s} r_{m2,\text{SF}s}} e^{2i\beta_{m,\text{SF}s} h_m} L^{-2/m,\text{sy}} \chi_{y\beta\gamma}
\end{aligned} \tag{11b}$$

### 透過 SFG

$$\begin{aligned}
E_{2p}^{+}(h_m^{+}) &= \sum_{\beta\gamma} E_{\text{vis}\beta}^0 E_{\text{ir}\gamma}^0 e \frac{t_{1m,\text{vis}\beta} t_{1m,\text{ir}\gamma} (1+r_{m2,\text{vis}\beta}) (1+r_{m2,\text{ir}\gamma}) e^{i(\beta_{m,\text{ir}} + \beta_{m,\text{vis}}) h_m}}{(1+r_{1m,\text{vis}\beta} r_{m2,\text{vis}\beta}) e^{2i\beta_{m,\text{vis}} h_m} (1+r_{1m,\text{ir}\gamma} r_{m2,\text{ir}\gamma}) e^{2i\beta_{m,\text{ir}} h_m}} \\
&\times \frac{(1+r_{m1,\text{SF}p} e^{2i\beta_{m,\text{SF}p} h_m}) L^{+2/m,\text{px}} \chi_{x\beta\gamma} + (-1+r_{m1,\text{SF}p} e^{2i\beta_{m,\text{SF}p} h_m}) L^{+2/m,\text{pz}} \chi_{z\beta\gamma}}{1+r_{1m,\text{SF}p} r_{m2,\text{SF}p}} e^{2i\beta_{m,\text{SF}p} h_m}
\end{aligned} \tag{12a}$$

$$\begin{aligned}
E^+_{2s}(h_m^+) &= \sum_{\beta, \gamma} E^0_{vis, \beta} E^0_{ir, \gamma} e^{\frac{t_{1m, vis, \beta} t_{1m, ir, \gamma} (1 + r_{m2, vis, \beta})(1 + r_{m2, ir, \gamma}) e^{i(\beta_{m, ir} + \beta_{m, vis})h_m}}{(1 + r_{1m, vis, \beta} r_{m2, vis, \beta} e^{2i\beta_{m, vis} h_m})(1 + r_{1m, ir, \gamma} r_{m2, ir, \gamma} e^{2i\beta_{m, ir} h_m})}} \\
&\times \frac{(1 + r_{m1, SF, s} e^{2i\beta_{m, SF} h_m}) L^+_{2/m, sy} \chi_{\beta, \gamma}}{1 + r_{1m, SF, s} r_{m2, SF, s} e^{2i\beta_{m, SF} h_m}}
\end{aligned} \tag{12b}$$

**膜バルク** [ $0 < z < h_m$ ,  $\mathbf{A}_{n=0,1,2} : \mathbf{x} = -2n h_m \tan \theta_{m, SF} - z \tan \theta_{m, SF}$ ,  $\mathbf{B}_{n=-1,0,1,2} : \mathbf{x} = -(2n+1) h_m \tan \theta_{m, SF} + z \tan \theta_{m, SF}$ ] からの SFG

### 反射 SFG

$$\begin{aligned}
E^-_{1p}(0^-) &= \frac{i}{\delta} \sum_{\beta, \gamma} E^0_{vis, \beta} E^0_{ir, \gamma} \\
&\times \frac{t_{m1, SF, p} t_{1m, vis, \beta} t_{1m, ir, \gamma}}{(1 + r_{1m, SF, p} r_{m2, SF, p} e^{2i\beta_{m, SF} h_m})(1 + r_{1m, vis, \beta} r_{m2, vis, \beta} e^{2i\beta_{m, vis} h_m})(1 + r_{1m, ir, \gamma} r_{m2, ir, \gamma} e^{2i\beta_{m, ir} h_m})} \\
&\times \left\{ \frac{1}{\beta_{m, SF} - \beta_{m, vis} - \beta_{m, ir}} \right. \\
&\quad \times \{ \chi_{x\beta\gamma} L^-_{1/m, px} [r_{m2, vis, \beta} r_{m2, ir, \gamma} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i(\beta_{m, vis} + \beta_{m, ir})h_m}) \\
&\quad + r_{m2, SF, p} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i\beta_{m, SF} h_m})] \\
&\quad + \chi_{x\beta\gamma} L^-_{1/m, pz} [r_{m2, vis, \beta} r_{m2, ir, \gamma} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i(\beta_{m, vis} + \beta_{m, ir})h_m}) \\
&\quad - r_{m2, SF, p} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i\beta_{m, SF} h_m})] \} \\
&+ \frac{1}{\beta_{m, SF} - \beta_{m, vis} + \beta_{m, ir}} \\
&\quad \times \{ \chi_{x\beta\gamma} L^-_{1/m, px} [r_{m2, vis, \beta} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i\beta_{m, vis} h_m}) \\
&\quad + r_{m2, SF, p} r_{m2, ir, \gamma} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i(\beta_{m, SF} + \beta_{m, ir})h_m})] \\
&\quad + \chi_{x\beta\gamma} L^-_{1/m, pz} [r_{m2, vis, \beta} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i\beta_{m, vis} h_m}) \\
&\quad - r_{m2, SF, p} r_{m2, ir, \gamma} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i(\beta_{m, SF} + \beta_{m, ir})h_m})] \} \\
&+ \frac{1}{\beta_{m, SF} + \beta_{m, vis} - \beta_{m, ir}} \\
&\quad \times \{ \chi_{x\beta\gamma} L^-_{m/m, px} [r_{m2, ir, \gamma} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i\beta_{m, ir} h_m}) \\
&\quad + r_{m2, SF, p} r_{m2, vis, \beta} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i(\beta_{m, SF} + \beta_{m, vis})h_m})] \\
&\quad + \chi_{x\beta\gamma} L^-_{m/m, pz} [r_{m2, ir, \gamma} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i\beta_{m, ir} h_m}) \\
&\quad - r_{m2, SF, p} r_{m2, vis, \beta} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i(\beta_{m, SF} + \beta_{m, vis})h_m})] \} \\
&+ \frac{1}{\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir}} \\
&\quad \times \{ \chi_{x\beta\gamma} L^-_{m/m, px} [(e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - 1) \\
&\quad + r_{m2, SF, p} r_{m2, vis, \beta} r_{m2, ir, \gamma} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i(\beta_{m, vis} + \beta_{m, ir})h_m})] \\
&\quad + \chi_{x\beta\gamma} L^-_{m/m, pz} [(e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - 1) \\
&\quad - r_{m2, SF, p} r_{m2, vis, \beta} r_{m2, ir, \gamma} (e^{i(\beta_{m, SF} + \beta_{m, vis} + \beta_{m, ir})h_m} - e^{2i(\beta_{m, vis} + \beta_{m, ir})h_m})] \}
\end{aligned} \tag{13a}$$



$$\begin{aligned}
E_{1s}^{-}(0^{-}) &= \frac{i}{\delta} \sum_{\beta\gamma} E_{vis,\beta}^0 E_{ir,\gamma}^0 \\
&\times \frac{t_{m1,SF,s} t_{1m,vis,\beta} t_{1m,ir,\gamma}}{(1+r_{1m,SF,s} r_{m2,SF,s} e^{2i\beta_{m,SF} h_m})(1+r_{1m,vis,\beta} r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})} \\
&\times \left\{ \frac{1}{\beta_{m,SF} - \beta_{m,vis} - \beta_{m,ir}} \right. \\
&\quad \times \{ \chi_{y\beta\gamma} L_{1/m,SY}^{-} [r_{m2,vis,\beta} r_{m2,ir,\gamma} (e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i(\beta_{m,vis} + \beta_{m,ir}) h_m}) \\
&\quad + r_{m2,SF,s} (e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i\beta_{m,SF} h_m})] \\
&\quad + \frac{1}{\beta_{m,SF} - \beta_{m,vis} + \beta_{m,ir}} \\
&\quad \times \{ \chi_{y\beta\gamma} L_{1/m,SY}^{-} [r_{m2,vis,\beta} (e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i\beta_{m,vis} h_m}) \\
&\quad + r_{m2,SF,s} r_{m2,ir,\gamma} (e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i(\beta_{m,SF} + \beta_{m,ir}) h_m})] \\
&\quad + \frac{1}{\beta_{m,SF} + \beta_{m,vis} - \beta_{m,ir}} \\
&\quad \times \{ \chi_{y\beta\gamma} L_{m/m,SY}^{-} [r_{m2,ir,\gamma} (e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i\beta_{m,ir} h_m}) \\
&\quad + r_{m2,SF,s} r_{m2,vis,\beta} (e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i(\beta_{m,SF} + \beta_{m,vis}) h_m})] \\
&\quad + \frac{1}{\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}} \\
&\quad \times \{ \chi_{y\beta\gamma} L_{m/m,SY}^{-} [(e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - 1) \\
&\quad + r_{m2,SF,s} r_{m2,vis,\beta} r_{m2,ir,\gamma} (e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i(\beta_{m,vis} + \beta_{m,ir}) h_m})] \}
\end{aligned} \tag{13b}$$

### 透過 SFG

$$\begin{aligned}
E_{2p}^{+}(h_m^{+}) &= \frac{i}{\delta} \sum_{\beta\gamma} E_{vis,\beta}^0 E_{ir,\gamma}^0 \\
&\times \frac{t_{1m,vis,\beta} t_{1m,ir,\gamma} e^{i\beta_{m,SF} h_m}}{(1+r_{1m,SF,p} r_{m2,SF,p} e^{2i\beta_{m,SF} h_m})(1+r_{1m,vis,\beta} r_{m2,vis,\beta} e^{2i\beta_{m,vis} h_m})(1+r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir} h_m})} \\
&\times \{ L_{2/m,pX}^{+} \chi_{x\beta\gamma} \left[ \frac{[e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - 1] + r_{m1,SF,p} r_{m2,vis,\beta} r_{m2,ir,\gamma} [e^{2i(\beta_{m,vis} + \beta_{m,ir}) h_m} - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m}]}{\beta_{m,SF} - \beta_{m,vis} - \beta_{m,ir}} \right. \\
&\quad + \frac{r_{m2,ir,\gamma} [e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i\beta_{m,ir} h_m}] + r_{m1,SF,p} r_{m2,vis,\beta} [e^{2i\beta_{m,vis} h_m} - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m}]}{\beta_{m,SF} - \beta_{m,vis} + \beta_{m,ir}} \\
&\quad + \frac{r_{m2,vis,\beta} [e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i\beta_{m,vis} h_m}] + r_{m1,SF,p} r_{m2,ir,\gamma} [e^{2i\beta_{m,ir} h_m} - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m}]}{\beta_{m,SF} + \beta_{m,vis} - \beta_{m,ir}} \\
&\quad \left. + \frac{r_{m2,vis,\beta} r_{m2,ir,\gamma} [e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i(\beta_{m,vis} + \beta_{m,ir}) h_m}] + r_{m1,SF,p} [1 - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m}]}{\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}} \right] \\
&\quad + L_{2/m,pZ}^{+} \chi_{z\beta\gamma} \left[ \frac{[e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - 1] - r_{m1,SF,p} r_{m2,vis,\beta} r_{m2,ir,\gamma} [e^{2i(\beta_{m,vis} + \beta_{m,ir}) h_m} - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m}]}{\beta_{m,SF} - \beta_{m,vis} - \beta_{m,ir}} \right. \\
&\quad \left. + \frac{r_{m2,ir,\gamma} [e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m} - e^{2i\beta_{m,ir} h_m}] - r_{m1,SF,p} r_{m2,vis,\beta} [e^{2i\beta_{m,vis} h_m} - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}) h_m}]}{\beta_{m,SF} - \beta_{m,vis} + \beta_{m,ir}} \right]
\end{aligned}$$

$$\begin{aligned}
& + \frac{r_{m2,vis,\beta} [e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m} - e^{2i\beta_{m,vis}h_m}] - r_{m1,SF,p} r_{m2,ir,\gamma} [e^{2i\beta_{m,ir}h_m} - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m}]}{\beta_{m,SF} + \beta_{m,vis} - \beta_{m,ir}} \\
& + \frac{r_{m2,vis,\beta} r_{m2,ir,\gamma} [e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m} - e^{2i(\beta_{m,vis} + \beta_{m,ir})h_m}] - r_{m1,SF,p} [1 - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m}]}{\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}} \Big] \Big\} \quad (14a)
\end{aligned}$$

$$\begin{aligned}
E^+_{2s}(h_m^+) &= \frac{i}{\delta} \sum_{\beta,\gamma} E^0_{vis,\beta} E^0_{ir,\gamma} \\
& \times \frac{t_{1m,vis,\beta} t_{1m,ir,\gamma} e^{i\beta_{m,SF}h_m}}{(1 + r_{1m,SF,s} r_{m2,SF,s} e^{2i\beta_{m,SF}h_m})(1 + r_{1m,vis,\beta} r_{m2,vis,\beta} e^{2i\beta_{m,vis}h_m})(1 + r_{1m,ir,\gamma} r_{m2,ir,\gamma} e^{2i\beta_{m,ir}h_m})} \\
& \times L^+_{2/m, sy} \chi_{\beta\gamma} \Big\{ \frac{[e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m} - 1] + r_{m1,SF,s} r_{m2,vis,\beta} r_{m2,ir,\gamma} [e^{2i(\beta_{m,vis} + \beta_{m,ir})h_m} - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m}]}{\beta_{m,SF} - \beta_{m,vis} - \beta_{m,ir}} \\
& + \frac{r_{m2,ir,\gamma} [e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m} - e^{2i\beta_{m,ir}h_m}] + r_{m1,SF,s} r_{m2,vis,\beta} [e^{2i\beta_{m,vis}h_m} - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m}]}{\beta_{m,SF} - \beta_{m,vis} + \beta_{m,ir}} \\
& + \frac{r_{m2,vis,\beta} [e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m} - e^{2i\beta_{m,vis}h_m}] + r_{m1,SF,s} r_{m2,ir,\gamma} [e^{2i\beta_{m,ir}h_m} - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m}]}{\beta_{m,SF} + \beta_{m,vis} - \beta_{m,ir}} \\
& + \frac{r_{m2,vis,\beta} r_{m2,ir,\gamma} [e^{i(-\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m} - e^{2i(\beta_{m,vis} + \beta_{m,ir})h_m}] + r_{m1,SF,s} [1 - e^{i(\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir})h_m}]}{\beta_{m,SF} + \beta_{m,vis} + \beta_{m,ir}} \Big\} \quad (14b)
\end{aligned}$$