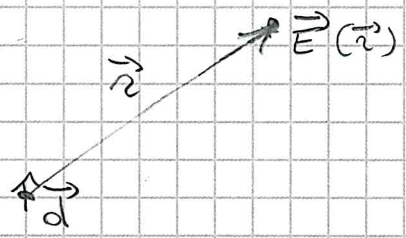
Optical reciprocity and optical chirality



dipole emission:

$$\vec{E}(\underline{r}) = \left(\frac{\omega}{c}\right)^2 \frac{e^{i\frac{\omega}{c}r}}{r^3}\left[(\underline{r}\times\underline{d})\times\underline{r}\right]$$

$$= \left(\frac{\omega}{c}\right)^2 \frac{e^{i\frac{\omega}{c}r}}{r^3}\left[r^2\underline{d} - \underline{r}(\underline{r}\cdot\underline{d})\right]$$

example

$\underline{d} = \frac{d_0}{\sqrt{2}}(\hat{e}_x + i\hat{e}_y)$ circular polarization

for $\underline{r} \in xy$ plane $\quad \underline{r} = r(\cos\theta, \sin\theta)$

$$E(\underline{r}) = -\frac{\omega^2}{c^2}\frac{e^{i\frac{\omega}{c}r}}{r^3}\left[\frac{d_0}{\sqrt{2}}(x+iy)\underline{r} - r^2\frac{d_0}{\sqrt{2}}(\hat{e}_x + i\hat{e}_y)\right]$$

$$= -\frac{\omega^2}{c^2}\frac{e^{i\frac{\omega}{c}r}}{r^3}\frac{d_0}{\sqrt{2}}\Big[\cancel{x^2\hat{e}_x} + xy\,\hat{e}_y + i\,xy\,\hat{e}_x + \cancel{i\,y^2\hat{e}_y} +$$
$$- (\cancel{x^2} + y^2)\hat{e}_x - (x^2 + \cancel{y^2})\,i\,\hat{e}_y\Big]$$

$$= -\frac{\omega^2}{c^2}\frac{e^{i\frac{\omega}{c}r}}{r^3}\frac{d_0}{\sqrt{2}}\left[x(y - ix)\hat{e}_y + y(ix - y)\hat{e}_x\right]$$

$$= \frac{\omega^2}{c^2}\frac{e^{i\frac{\omega}{c}r}}{r^3}\frac{d_0}{\sqrt{2}}\left[+i r^2\cos\theta\, e^{i\theta}\hat{e}_y - i\sin\theta\, e^{i\theta}\hat{e}_x\right]$$

$\boxed{\hat{e}_z \times \underline{r} = (\cos\theta\,\hat{e}_y - \sin\theta\,\hat{e}_x)r \quad \text{rotates by } 90^\circ \text{ ccw}}$

$$= i A \frac{e^{i\frac{\omega}{c}r}}{r^2}\frac{d_0}{\sqrt{2}}\underbrace{e^{i\theta}}_{\text{angle-dep. phase}}\underbrace{(\hat{e}_z\times\underline{r})}_{\text{rotated by } 90^\circ \text{ ccw}}$$

$+i\hat{e}_y \downarrow \qquad \circlearrowleft \qquad \uparrow -i\hat{e}_y$

<u>Susceptibility</u> $\overleftrightarrow{\chi} = (\chi_0 + \chi_1)|\sigma_+\rangle\langle\sigma_+| + (\chi_0 - \chi_1)|\sigma_-\rangle\langle\sigma_-|$

different response in $|\sigma_\pm\rangle = \frac{1}{\sqrt{2}}\left[|x\rangle \pm i|y\rangle\right]$

circular basis

$$\overleftrightarrow{\chi} = \begin{pmatrix} \chi_0 & -i\chi_1 \\ +i\chi_1 & \chi_0 \end{pmatrix}$$

$$\hat{e}_\theta = (\cos\theta, \sin\theta)$$

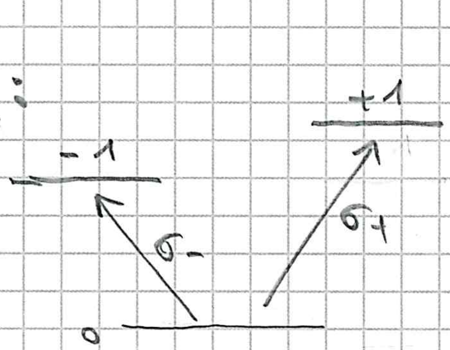
$$\overleftrightarrow{\chi}\,\hat{e}_\theta = \left(\chi_0\cos\theta - i\chi_1\sin\theta\right)\hat{e}_x + \left(i\chi_1\cos\theta + \chi_0\sin\theta\right)\hat{e}_y$$

$$= \bar{\chi}\left[\cos(\theta+\alpha)\,\hat{e}_x + \sin(\theta+\alpha)\,\hat{e}_y\right] = \hat{e}_{\theta+\alpha}\cdot\bar{\chi}$$

where $\chi_0 = \bar{\chi}\cos\alpha$; $\chi_1 = \bar{\chi}\sin\alpha$

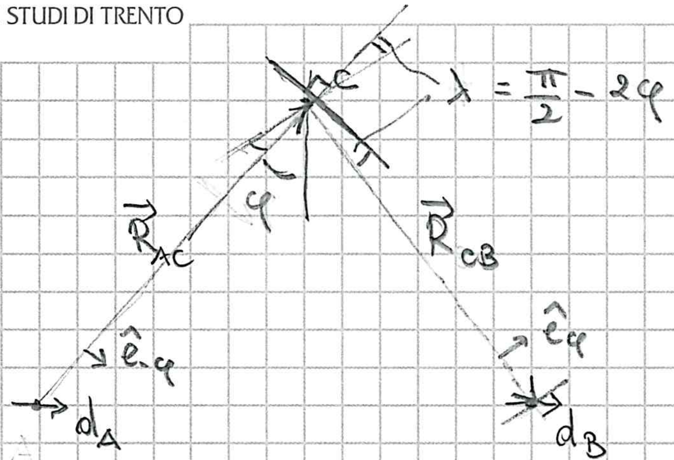
$\Rightarrow$ dipole rotated by α from $\vec{E}$.

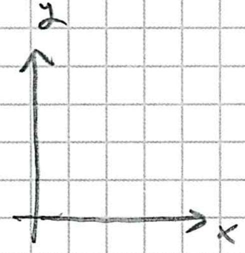
<u>Physical example</u>:



Zeeman-shifted transitions

(or, also, 2D Drude-Lorentz in magnetic field $\vec{B}$)





$\vec{d}_A = \vec{d}_B = d_0 \cdot \hat{e}_x$

No direct emission $A \to B$ nor $B \to A$.

Lowest order scattering : $A \to C \to B$

$B \to C \to A$

$|\vec{R}_{AC}| = |\vec{R}_{BC}| = R$

$A \to C \to B$)

$$\vec{E}(r_c) = \left(\frac{\omega}{c}\right)^2 \frac{e^{i\frac{\omega}{c}R}}{R^3} \left(\vec{R}_{AC} \times \vec{d}_A\right) \times \vec{R}_{AC}$$

$$\vec{E}(r_B) = \left(\frac{\omega}{c}\right)^2 \frac{e^{i\frac{\omega}{c}R}}{R^3} \left(\vec{R}_{CB} \times \vec{d}_c\right) \times \vec{R}_{CB}$$

with $\vec{d}_c = \overleftrightarrow{\chi}\, \vec{E}(r_c)$

$\vec{E}_c(r_c) \sim \cos\varphi \cdot \hat{e}_{-\varphi}$

$\vec{d}_c \sim \cos\varphi \cdot \hat{e}_{-\varphi+\alpha}$

polarization rotation

$\vec{E}(r_B) \sim \cos\varphi \cdot \hat{e}_{\varphi} \sin(\lambda + \alpha)$

B → C → A)

$$\vec{E}(r_C) \sim \cos\varphi \cdot \hat{e}_\varphi$$

$$\vec{d}_C \sim \cos\varphi \cdot \hat{e}_{\varphi+\alpha}$$

$$\vec{E}(r_A) \sim \cos\varphi \quad \hat{e}_{-\varphi} \sin(\lambda-\alpha)$$

* for $\varphi = \pi/4 \longrightarrow \lambda = 0$

$\Longrightarrow$ $\vec{E}(r_B)$ and $\vec{E}(r_A)$ have opposite phases

* for $\alpha = \lambda \longrightarrow \vec{E}(r_B) \neq 0$ but $\vec{E}(r_A) = 0$

$\Longrightarrow$ OPTICAL DIODE

transmission $A \to B$ but not $B \to A$

Generic scattering

$$\vec{E}(r_B) = \overleftrightarrow{G}_{CB} \cdot \vec{d}_C = \overleftrightarrow{G}_{CB} \cdot \overleftrightarrow{X} \cdot \vec{E}(r_C) = \overleftrightarrow{G}_{CB} \overleftrightarrow{X} \overleftrightarrow{G}_{AC} \vec{d}_A$$

$$\vec{E}(r_A) = \overleftrightarrow{G}_{CA} \overleftrightarrow{X} \overleftrightarrow{G}_{BC} \vec{d}_B$$

where $\overleftrightarrow{G}_{ij} = \left(\frac{\omega}{c}\right)^2 \frac{e^{i\frac{\omega}{c}R_{ij}}}{R_{ij}^3} \left[R_{ij}^2 \cdot \mathbb{1} - (\vec{R}_{ij} \cdot \mathbb{1}) \cdot \vec{R}_{ij} \right]$

NOTE: $\overleftrightarrow{G}_{ij} = \overleftrightarrow{G}_{ji}$ (even in R_{ij})

$$\left(\overleftrightarrow{G}_{ij}\right)_{\alpha\beta} = \left(\overleftrightarrow{G}_{ij}\right)_{\beta\alpha} \text{ for } \alpha,\beta = \hat{e}_x, \hat{e}_y\ \hat{e}_z$$

$\left[\left(\overleftrightarrow{G}_{ij}\right)_{\alpha\beta} = \langle e_\alpha | \overleftrightarrow{G}_{ij} | e_\beta \rangle\right]$ ↳ symmetric

(not for complex vectors, e.g. $\sigma_\pm$!)

If we assume $\left(\overleftrightarrow{X}\right)_{\alpha\beta} = \left(\overleftrightarrow{X}\right)_{\beta\alpha}$:

then $$\langle \hat{e}_\beta | G_{CB} X\, G_{AC} | \hat{e}_\alpha \rangle = \langle \hat{e}_\alpha | G_{CA} \cdot X\, G_{BC} | \hat{e}_\beta \rangle$$

which means <u>RECIPROCITY</u>

$\hat{e}_\beta$ component @ r_B from source @ r_A polarised $\hat{e}_\alpha$

=

$\hat{e}_\alpha$ component @ r_A from source @ r_B polarised $\hat{e}_\beta$

Zeeman-split transition $\overleftrightarrow{X}$ is <u>NOT</u> symmetric!

↳ reciprocity can be broken.