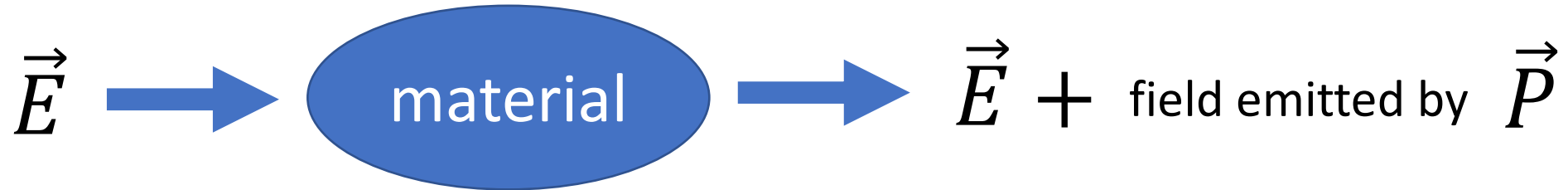


# Summary of NLO lecture #1

26.09.2018

# Polarization

- Plane wave hits material
- $\vec{E}$  couples to molecules in a medium
- $\vec{E}$  accelerates charge
- Accelerated charge emits light



Assumptions:

- 1) Plane wave
- 2) Dipole approximation

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P} + \dots$$

This doesn't mean we exclude non-linear effects.

# Polarization

- $\vec{P}$  is a function of  $\vec{E}$
- $\vec{P}$  can be expanded around  $\vec{E}$

$$\vec{P}(\vec{E}) = \vec{P}^{(0)}(\vec{E}) + \vec{P}^{(1)}(\vec{E}) + \vec{P}^{(2)}(\vec{E}) + \vec{P}^{(3)}(\vec{E}) + \dots$$

constant

linear

**non-linear**

- To invoke  $\vec{P}^{(2+)}$  we need a high EM field strength

# $\vec{P}$ can be complicated

- $\vec{P}$  can have different direction than  $\vec{E}$
- $\vec{P}$  can have multiple frequencies ( $\omega_1$  and  $\omega_2$  in,  $\omega_0$  out)
- $\vec{P}$  can be non-local ( $\vec{P}(\vec{b})$  depends on  $\vec{E}(\vec{a})$ )
- $\vec{P}$  can be temporally non-local ( $\vec{P}$  depends on  $\vec{E}$  in the past)

- 1) Describe  $\vec{P}$  (with some of these complications)
- 2) Use description to define  $\chi^{(n)}$   
(non-linear optical susceptibility)

# Describe $\vec{P}$ with complications

$$P_i^{(1)}(r, t) = \epsilon_0 \int d\tau_1 \int dr_1 R_{ij}(\tau_1, r_1) E_j(t - \tau_1, r - r_1)$$

$$P_i^{(1)}(r, t) = \epsilon_0 \int d\tau_1 R_{ijk}(\tau_1) E_j(t - \tau_1, r)$$

$$P_i^{(1)}(r, t) = \epsilon_0 \int d\tau_1 \int dr_1 R_{ijk}(\tau_1, r_1) E_j(t - \tau_1, r - r_1)$$

$$P_i^{(2)}(r, t) = \epsilon_0 \int d\tau_1 \int d\tau_2 \int dr_1 \int dr_2 R_{ijk}(\tau_1, r_1, \tau_2, r_2) E_j(t - \tau_1, r - r_1) E_k(t - \tau_2, r - r_2)$$

$$P_i^{(2)}(r, t) = \epsilon_0 \int d\tau_1 \int d\tau_2 R_{ijk}(\tau_1, \tau_2) E_j(t - \tau_1, r) E_k(t - \tau_2, r)$$

FT()

FT()

+ reordering...

Define  $\chi^{(n=2)}$

$$P_i^{(2)}(\omega_0) = \varepsilon_0 \chi_{ijk}^{(2)}(\omega_0; \omega_1; \omega_2) E_j(\omega_1) E_k(\omega_2)$$

$$\chi_{ijk}^{(2)}(\omega_0; \omega_1; \omega_2) = \int d\tau_1 \int d\tau_2 R_{ijk}^{(2)}(\tau_1, \tau_2) e^{i\omega(\tau_1 + \tau_2)}$$

... we have a way of defining a non-linear response without losing any generality.

# Remarks about units

- Different susceptibility orders have different units
- The higher the order, the lower the value
  
- $\chi^{(1)}$   $[-]$   $\sim 1$
- $\chi^{(2)}$   $\left[\frac{m}{V}\right]$   $\sim 1 \cdot 10^{-12}$
- $\chi^{(2)}$   $\left[\frac{m^2}{V^2}\right]$   $\sim 1 \cdot 10^{-24}$
- ...

# Remarks about macro- vs micro-scopic $\vec{P}$

$$\vec{P} = \sum_i \vec{p}_i = N \cdot \langle \vec{p}_i \rangle$$

$$\vec{p}_i = \alpha_{ij}^{(1)} E_j + \beta_{ijk}^{(2)} E_j E_k + \beta_{ijkl}^{(3)} E_j E_k E_l + \dots$$

- N – number of molecules
- $\vec{p}_i$  – atomic/molecular polarization
- $\langle \rangle$  – spatial average

Messy convention:

1)  $\alpha^{(1)}, \alpha^{(2)}, \alpha^{(3)}, \dots$

2)  $\alpha^{(1)}, \beta^{(2)}, \beta^{(3)}, \dots$

3)  $\alpha, \beta, \gamma, \dots$

N-th order (hyper)polarizability