Non linear Optics - Summary of lesson 5

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AHO Model

- HO model for the atom

Lorentz: explanation of how an electromagnetic field would interact with gases.

Use it with:

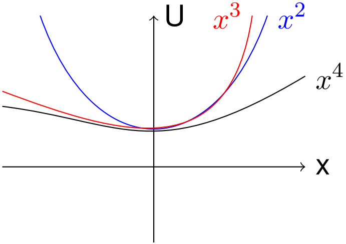
- dielectric constant/dispersion
- the resonance frequency of a plasma
- simple resonance phenomena

AHO Model

- When does the AHO work?
  - Gases
  - Simple regular non-mettalic solids
  - cases where $\omega < \omega_0$, i.e. off resonance

AHO on hydrogen atom

- Forces on an electron, attached to its nucleus:
  - driving force
  - spring force (Hooke's Law)
  - damping term (F   velocity)
  - Newton



- The energy of the system:

$$U(x) = U(0) + \left(\frac{\partial U}{\partial x}\right)\bigg|_{x=0} x + \frac{1}{2}\left(\frac{\partial^2 U}{\partial x^2}\right) x^2 + \frac{1}{6}\left(\frac{\partial^3 U}{\partial x^3}\right) x^3 + \frac{1}{24}\left(\frac{\partial^4 U}{\partial x^4}\right) x^4$$

AHO on hydrogen atom

- Force components:
  - $F = -\frac{\partial U}{\partial x}$
  - $\widetilde{F_N} = m\frac{d^2x}{dt^2}$
  - $\widetilde{F_b} = -m\omega_0 x - max^2 + mbx^3$
  - $\widetilde{F_d} = -2\gamma m\frac{d\widetilde{x}}{dt}$
  - $\widetilde{F_d} = -e\widetilde{E}(t)$
- Solve:

$$\boxed{\ddot{\widetilde{x}} + 2\gamma\dot{\widetilde{x}} + \omega_0^2\widetilde{x} + a\widetilde{x}^2 - b\widetilde{x}^3 = -e\frac{\widetilde{E}(t)}{m}}$$

Centrosymmetric/Isotropic Model

- The binding potential changes slightly

$$U(\widetilde{x}) = \frac{1}{2}m\omega_0\widetilde{x}^2 - \frac{1}{4}mb\widetilde{x}^4$$

With $\vec{x}$ it gets:

$$F_{bind} = -m\omega_0(\vec{x}\cdot\vec{x}) + mb\vec{x}(\vec{x}\cdot\vec{x})$$

$$\vec{E} = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t} + \ c.c.$$

$$\longrightarrow F_{driving} = -\frac{e}{m}\sum E(\omega_i)e^{-i\omega_i t}$$

Solving for $\vec{x}$

- We use a *perturbative* approach:

$$E \longrightarrow \lambda \cdot E \quad \lambda \in [0,1]$$

at the end $\lambda \longrightarrow 1$

We get:

$$\vec{x} = \lambda^1 \vec{x}^{(1)}(t) + \lambda^2 \vec{x}^{(2)}(t) + \lambda^3 \vec{x}^{(3)}(t) + ...$$

Decomposed:

$$\begin{aligned}
\lambda \ddot{\vec{x}}^{(1)} + \lambda^2 \ddot{\vec{x}}^{(2)} + \lambda^3 \ddot{\vec{x}}^{(3)} + \\
2\gamma(\lambda \dot{\vec{x}}^{(1)} + \lambda^2 \dot{\vec{x}}^{(2)} + \lambda^3 \dot{\vec{x}}^{(3)} + ...) + \\
\omega_0^2(\lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + ...) - \\
b(\lambda^3(\vec{x}^{(1)} \cdot \vec{x}^{(1)})\vec{x}^{(1)} + ...) = -\frac{\lambda e}{m}\vec{E}(t)
\end{aligned}$$

Solving for $\vec{x}$

- Group:

$$
\begin{array}{ll}
\lambda & \ddot{\vec{x}}^{(1)} + 2\gamma\dot{\vec{x}}^{(1)} + \omega_0^2 x^{(1)} = -\frac{eE(t)}{m} \\
\lambda^2 & \ddot{\vec{x}}^{(2)} + 2\gamma\dot{\vec{x}}^{(2)} + \omega_0^2 x^{(2)} = 0 \quad \longleftarrow \text{ no driving force} \\
\lambda^3 & \ddot{\vec{x}}^{(3)} + 2\gamma\dot{\vec{x}}^{(3)} + \omega_0^2 x^{(3)} = b(\vec{x}^{(1)} \cdot \vec{x}^{(1)})\vec{x}^{(1)} \quad (*)
\end{array}
$$

- Solution for $x^{(1)}$

$$
x^{(1)}(t) = \sum_n x^{(1)}(\omega_n) e^{-i\omega_n t} \quad (+c.c.)
$$

Solving for $x^{(1)}$

- After filling in:

$$\vec{x}^{(1)}(\omega_n) = \frac{-e\vec{E}(\omega_n)}{(\omega_0^2 - \omega_n^2 - 2i\gamma\omega_n)m} = \frac{-e\vec{E}(\omega_n)}{D(\omega_n)m}$$

$\longrightarrow$ linear polarization: $\vec{p}^{(1)}(t) = -e\vec{x}^{(1)}(t) \quad P^{(1)}(t) = Np^{(1)}(t)$

- Absorption/Emission has a Lorentzian lineshape: $I \sim |P^{(1)}|^2$
- Linear susceptibility

$$P_i^{(1)} = \epsilon_0 \chi_{ij}^{(1)} E_{ij}$$

$$\chi_{ij}^{(1)} = \frac{-Ne^2}{\epsilon_0 m D(\omega_n)} \delta_{ij}$$

Solution for $\vec{x}^2(t) \quad (\lambda^2)$

Without a driving force: there is no displacement!

$$\left.\begin{aligned} \vec{x}^{(2)} &= 0 \\ \vec{P}^{(2)} &= 0 \\ \chi^{(2)}_{ijk} &= 0 \end{aligned}\right\} \quad \text{agrees with our symmetry requirement.}$$

Solution for $\vec{x}^3(t) \quad (\lambda^3)$

- Driving:

$$b(\vec{x}^{(1)} \cdot \vec{x}^{(1)})\vec{x}^{(1)} = \frac{-be^3}{m^3} \sum_{n,m,p} \frac{\left[\vec{E}(\omega_n)\vec{E}(\omega_m)\right] \vec{E}(\omega_p)}{D(\omega_n)D(\omega_m)D(\omega_p)}$$

Constraint : $\omega_q = \omega_p + \omega_m + \omega_n$

- Form of solution:

$$x^{(3)}(t) = \sum_q x^{(3)}(\omega_q) e^{-i\omega_q t}$$

- Filling in in $(*)$:

$$\vec{x}^{(3)}(\omega_q) = \frac{-be^3}{m^3} \sum_q \frac{\left[\vec{E}(\omega_n)\vec{E}(\omega_m)\right] \vec{E}(\omega_p)}{D(\omega_n)D(\omega_m)D(\omega_p)D(\omega_q)}$$

Third order susceptibility

$$\vec{P}^{(3)} = -Ne\vec{x}^{(3)}$$

$$\chi^{(3)}_{ijkl} = \frac{P^{(3)}}{\epsilon_0 E_j E_k E_l}$$

Constraints on the number of elements: isotropic C_2 axis.
$\Longrightarrow$ a double occurance of each x,y,z elements (xxyy OK, not xyzz)
This can be arranged by using δ symbols:
$\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}$ can be used as multipliers.

$$P_i^{(3)} = \epsilon_0 \sum_{jikl} \sum_{nmp} \chi^{(3)}_{ijkl}(\omega_q; \omega_n, \omega_m, \omega_p) E_j(\omega_n) E_k(\omega_m) E_l(\omega_p)$$

Third order susceptibility

$$\left.\begin{aligned} \chi^{(3)}_{ijkl} &= \frac{Nbe^4[\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}]}{3\epsilon_0 m^3 D(\omega_n)D(\omega_m)D(\omega_p)D(\omega_q)} \\ \chi^{(1)}_{ijk} &= \frac{-Ne^2\delta_{ij}}{\epsilon_0 m D(\omega_n)} \end{aligned}\right\} \longrightarrow$$

$$\longrightarrow \chi^{(3)}_{ijkl} = \frac{bm\epsilon_0^3}{3N^3e^4}\left[\chi^{(1)}(\omega_q)\chi^{(1)}(\omega_p)\chi^{(1)}(\omega_n)\chi^{(1)}(\omega_m)\right]\cdot f$$

With : $f = \delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}$

Product of $\chi^{(1)\prime}s$!