Tutorial 2

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PH 421: Photonics
Due: 12:30 Thursday, August 24, 2023

Problem 2.1. Talking numbers: nonlinear dipole moments and polarization
A laser beam of frequency ω carrying 1 W of power is focused to a spot size of 30 µm diameter in a crystal having a refractive index of $n = 2$ and a second-order susceptibility of $\chi^{(2)} = 4 \times 10^{-11}$ m/V. Calculate numerically the amplitude $P(2\omega)$ of the component of the nonlinear polarization oscillating at frequency 2ω. Estimate numerically the amplitude of the dipole moment per atom $\mu(2\omega)$ oscillating at frequency 2ω. Compare this value with the atomic unit of dipole moment (ea_0, where a_0 is the Bohr radius) and with the linear response of the atom, that is, with the component $\mu(\omega)$ of the dipole moment oscillating at frequency ω.

Problem 2.2. Estimating absorption coefficient and refractive index
Perform an estimate of the magnitude of the on-resonance absorption coefficient of a dense atomic vapor assuming that the atomic number density is $N = 10^{17}$ cm^{-3}, that $\mu = 2.5ea_0$, that the transition vacuum wavelength is 0.6 µm, and that the transition is homogeneously broadened with a linewidth (FWHM) of 10 GHz. Under the same conditions, calculate the maximum value of the real part of the refractive index near the peak of the absorption line.

Problem 2.3. Intrinsic permutations and second order susceptibility
Please write out all the terms of the second order polarization expression and convince yourself that the choice of averaging the susceptibility expression for all combinations of the two input frequencies makes sense.

Problem 2.4. Second harmonic generation with free electrons
(a) Consider an electron in vacuum, under the action of a monochromatic light field of frequency ω, moving in the z direction. Light is polarized in the x direction. Calculate the nonlinear polarizability corresponding to second harmonic generation.
(b) Based on symmetry arguments, explain why we can get a second harmonic although the electron in vacuum follows inversion symmetry.

Problem 2.5. Nonlinear second order susceptibility: classical versus quantum
Please identify the differences in the quantum expression for the susceptibility versus the classical one.

Problem 2.6. Third order correction to the electronic wavefunction
(a) Please provide an expression for the third order correction to the electronic wavefunction in terms of eigenergies of the unperturbed Hamiltonian and the matrix element of the dipole operator in the basis of the eigenstates of the unperturbed Hamiltonian.
(b) Provide an expression for the third order nonlinear optical susceptibility using your result in part (a).

Problem 2.7. Frequency domain version of the wave equation in a nonlinear medium
Please derive the frequency domain version of the wave equation for the electric field in a nonlinear medium. You can leave the answer in terms of the linear dielectric function and the nonlinear polarization.

Q1)

$I = \frac{1W}{Area} = 2c\varepsilon_0 n|E|^2$

$\boxed{\text{Calculate } |E|^2}$

Now $P(2\omega)$ will only be contributed by $\chi^2 E^2$ $\therefore$ $P(2\omega) = \varepsilon_0 \chi^{(2)}(2\omega) E^2$

$\boxed{P(2\omega) = \varepsilon_0 \times 4 \times 10^{-11} \times |E|^2}$

$\mu(2\omega) N = P(2\omega)$

$\boxed{\mu(2\omega) = \frac{P(2\omega)}{N}}$, $\mu(\omega) = \frac{P(\omega)}{N}$

$P(\omega) = \varepsilon_0 \chi^{(1)} \times E = \varepsilon_0 (n^2-1) E$

$\boxed{\therefore \mu(\omega) = \frac{\varepsilon_0 (n^2-1) E}{N} \text{, where } N \approx 10^{28}}$

Q2

Dipole moment

$$\bar{p}^{(1)} = \langle \psi^{(0)} | \hat{\mu} | \psi^{(1)} \rangle + \langle \psi^{(1)} | \hat{\mu} | \psi^{(0)} \rangle$$

Since $\psi^{(1)} = \sum_m a_m^{(1)} u_m e^{-i\omega_m t}$

and $\psi^{(0)} = u_g e^{-i\omega_g t}$

$$\Rightarrow \bar{p}^{(1)} = \sum_{p,m} \left[\frac{\bar{\mu}_{gm} [\bar{\mu}_{mg} \cdot \bar{E}(\omega_p)] e^{-i\omega_p t}}{\hbar(\omega_{mg} - \omega_p)} + \frac{\bar{\mu}_{mg} [\bar{\mu}_{mg} \cdot \bar{E}(\omega_p)]^* e^{+i\omega_p t}}{\hbar(\omega_{mg}^* - \omega_p)} \right] \quad (4)$$

Let's assume a damping:

$$\omega_{mg} \rightarrow \omega_{mg} - i\Gamma_m/2$$

Converting the summation in eq. (4) to $e^{-i\omega_p t}$ only:

$$p^{(1)} = \sum_{p,m} e^{-i\omega_p t} \left[\frac{\bar{\mu}_{gm} \cdot [\bar{\mu}_{mg} \cdot \bar{E}(\omega_p)]}{\hbar(\omega_{mg} - \omega_p - i\Gamma_m/2)} + \frac{\bar{\mu}_{mg} [\bar{\mu}_{gm} \cdot \bar{E}(\omega_p)]}{\hbar(\omega_{mg} + \omega_p + i\Gamma_m/2)} \right]$$

Now we can easily calculate Susceptibility:

Since $P^{(1)}(\omega_p) = N p^{(1)}(\omega_p) = \epsilon_0 \chi^{(1)}(\omega_p) \cdot \bar{E}(\omega_p)$

$$\Rightarrow \chi^{(1)}(\omega) = \frac{N}{\epsilon_0 \hbar} \sum_m \frac{\bar{\mu}_{gm} \bar{\mu}_{mg}}{\omega_{mg} - \omega - i\Gamma_m/2} + \underbrace{\frac{\bar{\mu}_{mg} \bar{\mu}_{gm}}{\omega_{mg} + \omega + i\Gamma_m/2}}$$

Anti-resonant term

$$\Rightarrow \chi^{(1)}(\omega) = \frac{N}{\epsilon_0 \hbar} \sum_m \frac{\bar{\mu}_{gm}\bar{\mu}_{mg}}{\omega_{mg}-\omega-i\Gamma_m/2} + \underbrace{\frac{\bar{\mu}_{mg}\bar{\mu}_{gm}}{\omega_{mg}+\omega+i\Gamma_m/2}}$$

anti-resonant term
not important near
$\omega = \omega_{mg}$

According to the problem, we are close to a single transition ($\lambda_0 = 0.6\mu m$). So there will be only a single dominant term in the summation. Further, let's assume "isotropic" χ:

$$\Rightarrow \chi^{(1)} = \frac{N}{\epsilon_0 \hbar} \frac{|\mu|^2/3}{\omega_0 - \omega - i\Gamma_m/2}$$

orientational average

(1) Absorption coefficient at resonance

With $\omega = \omega_0$:

$$\chi^{(1)} = i\left[\frac{N}{\epsilon_0 \hbar} \frac{|\mu|^2/3}{(\Gamma_m/2)}\right]$$

Complex refractive index

$$\tilde{n} = n + iK = \sqrt{1+\chi^{(1)}}$$

$$I = I_0 e^{-\alpha L} = I_0 e^{-2k_0 K L}$$

$$\Rightarrow \text{absorption Coeff} = 2k_0 K = \alpha$$

(2) Max value of real part of refractive index

Note how $\chi^{(1)}$ looks

$$Re(\chi^{(1)}) = \frac{N|\mu|^2}{3\epsilon_0\hbar} \frac{\omega_0-\omega}{(\omega_0-\omega)^2+(\Gamma_m/2)^2}$$

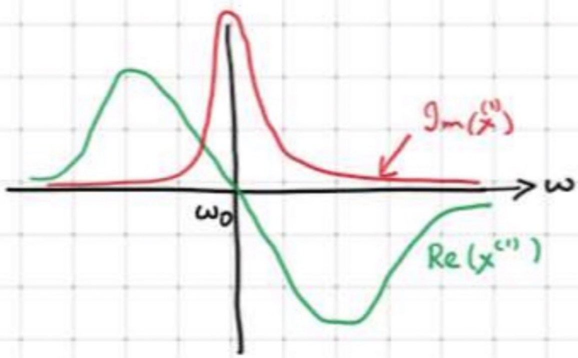
This is a function of the form:

$$f(x) = \frac{x}{x^2+a^2} \Rightarrow f'(x) = \frac{x^2+a^2-2x^2}{(\quad)^2}$$

$$\Rightarrow \quad x = \pm a$$

Whereas

$$Im(\chi^{(1)}) = \frac{N|\mu|^2}{3\epsilon_0\hbar} \frac{\Gamma_m/2}{(\omega_0-\omega)^2+(\Gamma_m/2)^2}$$



$\Rightarrow$ Max real part of refractive index:

$$n = \left[1 + \chi^{(1)}(\omega = \omega_0 - \Gamma_m/2)\right]^{\frac{1}{2}}$$

$$= 1.2$$

Imag is close to zero

$\square$

Q 3) $P_i^{(2)} = \varepsilon_0 \sum_{jk} \sum_{(pq)} \chi_{ijk}^{(2)}(\omega_p+\omega_q, \omega_q, \omega_p) E_j(\omega_q) E_k(\omega_p)$

$$= \varepsilon_0 \sum_{jk} \sum_{pq} \left[\chi_{ijk}^{(2)}(\omega_p+\omega_q, \omega_q, \omega_p) + \chi_{ikj}(\omega_p+\omega_q, \omega_p, \omega_q) \right] E_j(\omega_q) E_k(\omega_p)$$

So, now permutation symmetry (that involves averaging the susceptibility) is only a choice just for sake of calculation.

$\because$ we have only one linear eq^n

$$\chi_{ijk}^{(2)}(\omega_p+\omega_q, \omega_q, \omega_p) + \chi_{ikj}(\omega_p+\omega_q, \omega_p, \omega_q) = C$$

We have infinite set of choices, best is to take $\frac{C}{2}, \frac{C}{2}$. (There could be other choices such as $(C, 0)$, $(0, C)$, etc.)

Q^n 5) Quantum has summation over different frequencies (over all excited states) while classical case has one term involving only relevant ω's.

Q4) Lorentz Force $= -e[\vec{E} + \vec{v} \times \vec{B}]$

a) $\vec{v} = \hat{v}_x \hat{x} + \hat{v}_z \hat{z}$

$$= -e\left[\vec{E} + (v_x \hat{x} + v_z \hat{z}) \times \frac{E}{c} \hat{y}\right]$$

$$= -e\left[E\hat{x} + \frac{v_x E}{c} \hat{z} - \frac{v_z E}{c} \hat{x}\right]$$

$$= -eE\left[\hat{x}\left(1 - \frac{\dot{z}}{c}\right) + \frac{\dot{x}}{c} \hat{z}\right]$$

$$\therefore m\ddot{r} = -eE\left[\hat{x}\left(1 - \frac{\dot{z}}{c}\right) + \frac{\dot{x}}{c} \hat{z}\right]$$

$$= m(\ddot{x} + \ddot{z}) \qquad (\because \dot{y} = 0)$$

$$\left[\vec{E} = E_0 e^{-i\omega t} + E_0^* e^{i\omega t}\right]$$

$$x = \lambda x^{(1)} + \lambda^2 x^{(2)} + \ldots, \quad z = \lambda z^{(1)} + \lambda^2 z^{(2)} + \ldots$$

$$\lambda \equiv \quad m(\ddot{x}^{(1)} + \ddot{z}^{(1)}) = -e(E_0 e^{-i\omega t} + E_0^* e^{i\omega t})\hat{x}$$

$$x^{(1)} = \frac{e}{m\omega^2}(E_0 e^{-i\omega t} + E_0^* e^{i\omega t})$$

$z^{(1)} = 0$ [can be taken z_0, but using gauge $z_0 = 0$]

$$\lambda^2 \equiv m[\ddot{x}^{(2)} + \ddot{z}^{(2)}] = eE\left[\hat{x}\frac{\dot{z}^{(1)}}{c} - \frac{\dot{x}^{(1)}}{c}\hat{z}\right]$$

$$m\ddot{x}^{(2)} = \frac{e[E_0 e^{-i\omega t} + E_0^* e^{+i\omega t}]}{c}\,\dot{z} = 0$$ } solve for $\ddot{x}$

$$m\ddot{z}^{(2)} = -\frac{e(E_0 e^{-i\omega t} + E_0^* e^{+i\omega t}]}{c}\,\frac{e\,i\omega}{m\omega^2}[E_0^* e^{i\omega t} - E_0 e^{-i\omega t}]$$

$$m\ddot{z}^{(2)} = -\frac{e^2 i}{cm\omega}[E_0^{*2} e^{i2\omega t} - E_0^2 e^{-i2\omega t}] + \ldots \text{term}$$

→ Solve for $\ddot{z}$

$$m\ddot{z}^{(2)} = \cdot f(2\omega) + \ldots$$

Polarizability corresponding to second harmonic generation

$$\Rightarrow \quad Ne[\underbrace{x^{(2)}(2\omega)}_{=0}\hat{x} + z^{(2)}(2\omega)\hat{z}]$$

$= 0 \because x^{(2)}$ has not (2ω) term

$$\Rightarrow \quad Ne[z^{(2)}(2\omega)\hat{z}]$$

b)

Q^n] 6 Given in Ch-3 of Boyde

$Q^n 7)$ For a charge free & current free

$$\vec{\nabla}\cdot\vec{D} = \rho = 0$$

$$\nabla\cdot B = 0$$

$$\vec{\nabla}\times\vec{E} = -\frac{\partial B}{\partial t} \quad \ldots (iii)$$

$$\nabla\times H = \frac{\partial D}{\partial t} + J = \frac{\partial D}{\partial t}$$

Considering non-mag^n material $B = \mu_0 \vec{H}$

In eq^n (3)

$$\nabla\times(\nabla\times\vec{E}) = -\frac{\partial}{\partial t}(\vec{\nabla}\times\vec{B}) = -\frac{\partial}{\partial t}\mu_0(\vec{\nabla}\times\vec{H})$$

$$\nabla\times(\nabla\times\vec{E}) = -\mu_0\frac{\partial^2 D}{\partial t}$$

$$\therefore \nabla\times(\nabla\times\vec{E}) + \mu_0\frac{\partial^2 D}{\partial t} = 0$$

$$\boxed{(\because D = \epsilon_0\vec{E} + P)}$$

$$\Rightarrow \nabla\times(\nabla\times\vec{E}) + \mu_0\epsilon_0\frac{\partial^2 E}{\partial t} + \mu_0\frac{\partial P}{\partial t} = 0$$

$$\Rightarrow \underbrace{\nabla(\nabla\cdot E)}_{\text{is taken to zero or negligible}} - \nabla^2 E = \nabla\times(\nabla\times\vec{E})$$

$$\therefore \quad -\nabla^2 \vec{E} + \mu_0 \varepsilon_0 \frac{\partial^2 E}{\partial t} = -\mu_0 \frac{\partial P}{\partial t}$$

$$\Rightarrow \quad \nabla^2 \tilde{E} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \tilde{E} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 P}{\partial t^2}$$

or

$$\nabla^2 E - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 D}{\partial t^2} = 0$$

It is convinient to split into linear & non-linear part

$$P = P^{(1)} + P^{NL}$$

$$D = D^{(1)} + P^{NL}$$

where $\quad D^{(1)} = \varepsilon_0 D + P^{(1)}$

$$\therefore \quad \nabla^2 E - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 D^{(1)}}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial P^{NL}}{\partial t^2}$$

In general

$$\tilde{\mathbf{E}}_n(\mathbf{r}, t) = \mathbf{E}_n(\mathbf{r}) e^{-i\omega_n t} + \text{c.c.},$$
$$\tilde{\mathbf{D}}_n^{(1)}(\mathbf{r}, t) = \mathbf{D}_n^{(1)}(\mathbf{r}) e^{-i\omega_n t} + \text{c.c.},$$
$$\tilde{\mathbf{P}}_n^{\text{NL}}(\mathbf{r}, t) = \mathbf{P}_n^{\text{NL}}(\mathbf{r}) e^{-i\omega_n t} + \text{c.c.}$$

$$D_n^{(1)} = \varepsilon_0 \, \varepsilon^{(1)}(\omega_n) \, E_n(\omega_n)$$

$$\nabla^2 \tilde{E}_n(\omega_n) - \frac{\varepsilon^{(1)}(\omega_n)}{c^2} \frac{\partial^2 \tilde{E}_n}{\partial t^2} = \frac{1}{\varepsilon_0 c^2} \frac{\partial \tilde{P}_n^{NL}}{\partial t^2}$$

$$\Downarrow$$

$$\nabla^2 \mathbf{E}_n(\mathbf{r}) + \frac{\omega_n^2}{c^2} \boldsymbol{\epsilon}^{(1)}(\omega_n) \cdot \mathbf{E}_n(\mathbf{r}) = -\frac{\omega_n^2}{\epsilon_0 c^2} \mathbf{P}_n^{\mathrm{NL}}(\mathbf{r}).$$

Ans