Week 03 – problem set

Nonlinear Optics for Quantum Technologies

March 6th, 2025

1 The anharmonic (nonlinear) Lorentz oscillator model

This exercise is to be done at home before Thursday's lecture.

We will extend the Lorentz oscillator model to account for nonlinear response. Thus, we assume that the electrons bound to the nucleus are subjected to an anharmonic potential. We assume that the electric field is polarized along a principal axis x, causing a displacement of the oscillator along the $x-$direction. Around the equilibrium position $x = 0$, the potential can be expanded as a Taylor series: [1]

$$V(x) = \frac{1}{2}m\omega_0^2 x^2 + \frac{1}{3}m\beta_2 x^3 + \frac{1}{4}m\beta_3 x^4 \tag{1}$$

The prefactors are introduced so that the force exerted on the electrons is:

$$F(x) = -\frac{dV(x)}{dx} = -m\omega_0^2 x - m\beta_2 x^2 - m\beta_3 x^3 \tag{2}$$

1.1 Non-centrosymmetric medium

In a crystal lacking an inversion center, the lowest order nonlinearity is usually the cubic term in the potential (quadratic term in the restoring force). We remind the electron's equation of motion in this case:

$$m\ddot{x} + m\omega_0^2 x(t) + m\beta_2 x(t)^2 + m\gamma_0 \dot{x} = qE(t) \tag{3}$$

For small displacements x, we can consider the potential to be weakly anharmonic, i.e., $|\beta_2| x \ll \omega_0^2$. We will therefore solve the problem using a classical perturbation approach. We introduce a perturbation parameter [2] λ into the solution ansatz for $x(t)$,

$$x(t) = x_1(t) + \lambda x_2(t)$$

$$V(x) = \frac{1}{2}m\omega_0^2 x^2 + \lambda\left(\frac{1}{3}m\beta_2 x^3\right)$$

where x_1 is the known exact solution of the linear problem for $\lambda = 0$ and x_2 is the nonlinear correction.

If we assume a drive at frequency ω, we can write the unperturbed solution $x_1(t) = \frac{1}{2}\left(X_1(\omega)\exp(-i\omega t) + c.c.\right)$ where *c.c.* denotes the complex conjugate and

$$X_1(\omega) = \frac{\varepsilon_0}{Nq}\chi^{(1)}(\omega)E$$

is the complex amplitude related to the drive by the susceptibility $\chi^{(1)}$ derived in Lecture 02. (We note N the volumic density of oscillators and q their individual charge).

i. Insert the ansatz for $x(t)$ and $V(x)$ into the differential equation (3) and find the equation satisfied by $x_2(t)$ by keeping only the terms of order 1 in λ.

[1] If the material has a center of symmetry (inversion symmetry) the coefficient β_2 vanishes and β_3 is the lowest order perturbation

[2] $\lambda \to 1$ turns the anharmonicity; $\lambda \to 0$ turns it off

ii. What new frequencies appear due to the term $x_1(t)^2$?

iii. To solve for $x_2(t)$, use the ansatz:

$$x_2(t) = X_2(\omega = 0) + \frac{1}{2}(X_2(2\omega)\exp(-i2\omega t) + c.c.) \tag{4}$$

and find the complex amplitudes $X_2(\omega = 0)$ and $X_2(2\omega)$ by collecting terms oscillating at the same frequency.

iv. Write down the expression of the nonlinear polarization $P^{(2)}(t)$ associated with the nonlinear correction $x_2(t)$ and show that

a. the term corresponding to optical rectification (induced static field) and noted $\chi^{(2)}(0; \omega, -\omega)$, is proportional to $|\chi^{(1)}|^2$

b. the term corresponding to second harmonic generation, noted $\chi^{(2)}(2\omega; \omega, \omega)$, is proportional to $\chi^{(1)}(2\omega) \cdot \left(\chi^{(1)}(\omega)\right)^2$

The universal nature of these proportionality relations was first experimentally noticed by Robert C. Miller in Appl. Phys. Lett. 5, 17–19 (1964).

1.2 (optional) Centrosymmetric medium

In a medium whose point group contains a center of inversion, such as silicon, diamond, glass, liquids, etc., we will see that the coefficient β_2 vanishes and β_3 is the lowest order perturbation to the harmonic potential. Apply the method above to this case and identify all the frequencies contained in the nonlinear polarization $P^{(3)}(t)$

2 Wave propagation in anisotropic media

This exercise will be treated in class on Thursday.

Second-order optical nonlinearities are mostly used in anisotropic materials such as birefringent crystals since they allow phase matching despite phase velocity dispersion. We consider the linear response of an anisotropic, homogeneous, and lossless dielectric medium. We chose the axes Ox, Oy, and Oz to coincide with the principal optics axes of the crystal so that the permittivity $\underline{\underline{\epsilon}}$ is a diagonal tensor.

$$D_i = \epsilon_{ii} E_i \quad \text{where} \quad D_i = \vec{D} \cdot \vec{u}_i \quad ; \quad i \in \{x, y, z\} \tag{5}$$

1. Derive the propagation equation from Maxwell equations in a homogeneous, lossless dielectric medium (real permittivity tensor) without magnetic response ($\mu = 1$) for linearly polarised plane waves of the form $\vec{E} = \hat{e} E_0 \exp(i\vec{k} \cdot \vec{r} - \omega t)$ (polarization unit vector $\hat{e}$ and wave vector $\vec{k}$).
2. Deduce which polarizations can propagate unaltered within the crystal (the normal modes) and derive the dispersion relations $|\vec{k}(\omega)|$ characterizing these polarizations, in the following cases:

   (a) in an isotropic medium for which:

   $$\epsilon = \epsilon_{xx} = \epsilon_{yy} = \epsilon_{zz}$$

   (b) in a uniaxial crystal with the extraordinary axis along z, for which:

   $$\epsilon_{xx} = \epsilon_{yy} = \epsilon_{\perp} \text{ and } \epsilon_{zz} = \epsilon_{\parallel}.$$

   For the second case, you may follow these steps

   i. restrict the study to wavevectors that are in the Oy, Oz plane (since x and y directions are equivalent)

   ii. rewrite the propagation equation for $\vec{k} = k_y \hat{e}_y + k_z \hat{e}_z = k \cos\theta \, \hat{e}_y + k \sin\theta \, \hat{e}_z$ and an arbitrary polarization, and project it on the three basis vectors to obtain 3 different equations

   iii. consider the first equation (projected onto Ox) for waves polarized along x (called "ordinary" waves); solve it to obtain the corresponding dispersion for such waves

   iv. consider the other two equations for a polarisation orthogonal to $\hat{e}_x$ and obtain a single 4^{th} order polynomial equation in k

   v. make the ersatz $k = \frac{\omega}{c} n_\theta$ and show that this last equation is solved for

   $$\frac{1}{n_\theta^2} = \frac{\cos\theta^2}{n_o^2} + \frac{\sin\theta^2}{n_e^2}$$

   Express n_o and n_e as a function of the permittivity tensor components.
3. Demonstrate that the energy of the extraordinary wave does not propagate along $\vec{k}$, but with an angle γ from $\vec{k}$. This angle γ is called the birefringent or *walk-off* angle. Show that γ is equal to the angle between $\vec{D}$ and $\vec{E}$. Derive an expression for γ.

Reminder of some vectorial identities:

$$\mathbf{a} \cdot (\mathbf{b} \times \mathbf{c}) = \mathbf{b} \cdot (\mathbf{c} \times \mathbf{a}) = \mathbf{c} \cdot (\mathbf{a} \times \mathbf{b})$$
$$\mathbf{a} \times (\mathbf{b} \times \mathbf{c}) = (\mathbf{a} \cdot \mathbf{c})\mathbf{b} - (\mathbf{a} \cdot \mathbf{b})\mathbf{c}$$
$$(\mathbf{a} \times \mathbf{b}) \cdot (\mathbf{c} \times \mathbf{d}) = (\mathbf{a} \cdot \mathbf{c})(\mathbf{b} \cdot \mathbf{d}) - (\mathbf{a} \cdot \mathbf{d})(\mathbf{b} \cdot \mathbf{c})$$
$$\nabla \times \nabla\psi = 0$$
$$\nabla \cdot (\nabla \times \mathbf{a}) = 0$$
$$\nabla \times (\nabla \times \mathbf{a}) = \nabla(\nabla \cdot \mathbf{a}) - \nabla^2\mathbf{a}$$
$$\nabla \cdot (\psi\mathbf{a}) = \mathbf{a} \cdot \nabla\psi + \psi\nabla \cdot \mathbf{a}$$
$$\nabla \times (\psi\mathbf{a}) = \nabla\psi \times \mathbf{a} + \psi\nabla \times \mathbf{a}$$
$$\nabla(\mathbf{a} \cdot \mathbf{b}) = (\mathbf{a} \cdot \nabla)\mathbf{b} + (\mathbf{b} \cdot \nabla)\mathbf{a} + \mathbf{a} \times (\nabla \times \mathbf{b}) + \mathbf{b} \times (\nabla \times \mathbf{a})$$
$$\nabla \cdot (\mathbf{a} \times \mathbf{b}) = \mathbf{b} \cdot (\nabla \times \mathbf{a}) - \mathbf{a} \cdot (\nabla \times \mathbf{b})$$
$$\nabla \times (\mathbf{a} \times \mathbf{b}) = \mathbf{a}(\nabla \cdot \mathbf{b}) - \mathbf{b}(\nabla \cdot \mathbf{a}) + (\mathbf{b} \cdot \nabla)\mathbf{a} - (\mathbf{a} \cdot \nabla)\mathbf{b}$$

A more complete description of light propagation in anisotropic crystals may be found in the following books:

- G.R. Fowles *Introduction to modern optics* Chapter 6.7
- M. Born, E. Wolf *Principles of Optics* Chapter XV
- B.E.A. Saleh, M.C. Teich *Fundamentals of Photonics*, Chapter 6.3
- A. Yariv, P. Yeh *Optical Waves in Crystals*, Chapter 4
- L.D. Landau, E.M. Lifshitz *Electrodynamics of Continuous Media*, Chapter XI