

Module 2 : Nonlinear Frequency Mixing

Lecture 7 : Optical Response Of Anharmonic Oscillator

Objectives

In this lecture we will look at

- Anharmonic oscillator model for nonlinear optical response of dielectrics.
- Miller's rule for Nonlinear susceptibility.

We have so far considered the optical properties of dielectric media in the linear approximation. However, linearity of the response is almost always an approximation – whatever the probe field. For example, we know that the elastic response becomes nonlinear at high stresses and the magnetic response is often nonlinear. Nonlinear electrical response of various electronic components is crucial for electronic circuits. So one should have expected that the optical response should also be nonlinear. We will now attempt to understand what parameters determine the magnitude of optical non linearity and then, the conditions under which the effects of nonlinearities would become significant or unavoidable.

First, we notice that the Lorentz model of the atom gave us an exactly linear response. But real atoms are not harmonic oscillators. In an atom the energy level spacing decreases $\sim 1/n^3$ as we go higher in energy whereas for the harmonic oscillator it remains constant. Moreover the Lorentz model does not allow for ionization of the atom. These two features of the real atom can be incorporated in the Lorentz model by making the oscillator weakly anharmonic. Let us represent the potential energy of an electron in the model atom by:

$$V = m \left(\frac{1}{2} \omega_0^2 r^2 - \frac{1}{3} \lambda r^3 - \frac{1}{4} \eta r^4 \right) \quad (7.1)$$

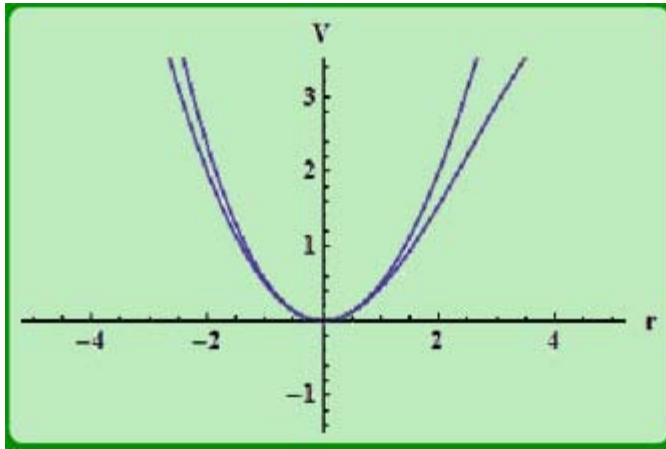


Figure 7.1. Anharmonic oscillator potential along with the corresponding harmonic approximation for small displacements

We note that this potential has a local minimum at the origin, and has two maxima at

$$r_{\pm} = -\frac{\lambda}{2\eta} \pm \sqrt{\frac{\lambda^2}{4\eta^2} + \frac{\lambda\omega_0^2}{\eta}}. \quad \text{This incorporates the finite ionization potential of a real atom in a crude way.}$$

As $r \rightarrow \pm\infty$, the potential goes to $-\infty$ but we will only consider electrons in the local minimum at $r = 0$.

We will now calculate the optical response of this model atom classically as before. (A quantum mechanical calculation would have to contend with the fact that the ground state in local well near $r \sim 0$ can also tunnel to $\pm\infty$ after sufficient time has lapsed). The classical equation of motion is

$$\ddot{r} + 2\gamma\dot{r} + \omega_0^2 r - \lambda r^2 - \eta r^3 = -(e/m)E \quad (7.2)$$

Even for the simple cosine time dependence of $E(t)$ this equation of motion is difficult to solve exactly. However, we notice that the magnitude of nonlinear terms is small, so long as the departure from

equilibrium is small which in turn is determined by the impressed field \mathbf{E} . This suggests that we should seek a power series solution in \mathbf{E} . We may then examine the solutions to see the limit of their range of validity.

Let

$$\mathbf{r} = \mathbf{r}_1 + \mathbf{r}_2 + \dots \quad (7.3)$$

Where \mathbf{r}_n is of the order \mathbf{E}^n . Substituting eq (7.3) in eq (7.2) and equating successively higher order in \mathbf{E} , we get:

$$\text{Order } E: \quad \ddot{\mathbf{r}}_1 + 2\gamma\dot{\mathbf{r}}_1 + \omega_0^2\mathbf{r}_1 = -(e/m)\mathbf{E} \quad (7.4)$$

$$\text{Order } E^2: \quad \ddot{\mathbf{r}}_2 + 2\gamma\dot{\mathbf{r}}_2 + \omega_0^2\mathbf{r}_2 = \lambda\mathbf{r}_1^2 \quad (7.5)$$

$$\text{Order } E^3: \quad \ddot{\mathbf{r}}_3 + 2\gamma\dot{\mathbf{r}}_3 + \omega_0^2\mathbf{r}_3 = 2\lambda\mathbf{r}_1\mathbf{r}_2 + \eta\mathbf{r}_1^3 \quad (7.6)$$

and so on. We have deliberately written these equations in a way which shows that they can be solved successively. The solution of Eq. (4) is already familiar. It may be rewritten as:

$$\mathbf{r}_1 = -(e/m)\mathbf{E}(\omega)\mathbf{F}(\omega)e^{-i\omega t} + \text{Complex Conjugate(C.C.)} \quad (7.7)$$

where $\mathbf{F}(\omega)$ is a convenient notation for the denominator.

$$\frac{1}{\omega_0^2 - 2i\gamma\omega - \omega^2} \equiv \mathbf{F}(\omega) \quad (7.8)$$

Substituting for \mathbf{r}_1 in Eq. (b), \mathbf{r}_2 is obtained as :

$$\mathbf{r}_2 = \frac{e^2}{m^2} \lambda \left\{ \mathbf{E}^2(\omega)\mathbf{F}^2(\omega)\mathbf{F}(2\omega)e^{-2i\omega t} + \mathbf{E}(\omega)\mathbf{E}^*(\omega)\mathbf{F}(\omega)\mathbf{F}^*(\omega)\mathbf{F}(0) \right\} + c.c. \quad (7.9)$$

Similarly,

$$\mathbf{r}_3 = -\frac{e^3}{m^3} \left[\left\{ (2\lambda^2\mathbf{F}(2\omega) + \eta)\mathbf{E}^3(\omega)\mathbf{F}^3(\omega)\mathbf{F}(3\omega) \right\} e^{-3i\omega t} + \left\{ (2\lambda^2\mathbf{F}(2\omega) + 4\lambda^2\mathbf{F}(0) + 3\eta)\mathbf{E}^2(\omega)\mathbf{E}^*(\omega)\mathbf{F}^3(\omega)\mathbf{F}^*(\omega) \right\} e^{-i\omega t} \right] + c.c. \quad (7.10)$$

Before examining those solutions closely, we note that the expansion (7.3) for \mathbf{r} automatically implies that the polarization vector $\mathbf{P} = -N e \mathbf{r}$ is also a power series in \mathbf{E} i.e. we may write:

$$\mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{(2)} + \dots \quad (7.11)$$

where $\mathbf{P}^{(n)}$ is of order n in \mathbf{E} . These solutions explicitly show several basic features of the optical response of nonlinear dielectrics; which can be established quite generally:

1. The linear response or \mathbf{P}_1 does not involve the anharmonic terms. However, even the response at ω is affected by the anharmonicity or optical nonlinearity of the medium. We notice for example, that $\mathbf{P}_3(\omega)$ has a resonance at $2\omega = \omega_0$ which gives rise to the nonlinear absorption called TWO PHOTON ABSORPTION. We will discuss this effect in greater detail later.
2. New frequencies may be involved in successively higher order response. The n^{th} order polarization has frequency component at all algebraic linear combinations of the n frequencies which must be contained in the impressed field. Thus, if the \mathbf{E} contains only the frequency ω , \mathbf{P}_2 will contain $\omega + \omega = 2\omega$ and $\omega - \omega = 0$. If, however, \mathbf{E} contained Fourier components at ω_1 as well as ω_2 , \mathbf{P}_2 will contain frequencies $2\omega_1$, $2\omega_2$, $\omega_1 \pm \omega_2$ and 0 .
3. Away from resonances, the various $\mathbf{F}(\omega)$ may be approximated by ω_0^{-2} [Strictly $\mathbf{F}(\omega) \leq \omega_0^{-2}$]

Then, we have:

$$\begin{aligned} \mathbf{r}_1 &= e\mathbf{E}/m\omega_0^2 \\ \mathbf{r}_2 &= \lambda e^2\mathbf{E}^2/m^2\omega_0^6 \\ \text{and } \mathbf{r}_3 &= \lambda^2 e^3\mathbf{E}^3/m^3\omega_0^{10} \text{ and } \sim \eta e^3\mathbf{E}^3/\omega_0^6 \end{aligned}$$

Now, for the power series expansion to be valid we should need that the amplitude is small such that for this amplitude, $\mathbf{r}_1 \sim e\mathbf{E}/m\omega_0^2$ is small such that for this amplitude, the terms $\lambda\mathbf{r}_1^2$ and $\eta\mathbf{r}_1^3$ in the restoring force should be small compared to the harmonic restoring force $\sim \omega_0^2\mathbf{r}$. When we get close to a resonance the validity of our expansion depends on the damping factor γ .

4. We may write polarization also as sum of various Fourier components. E.g.

$$P_1, P_2, P_3$$

$$P_2 = P_2(2\omega)e^{-2i\omega t} + P_2(0) + \text{Complex Conjugate}$$

Further, a second order susceptibility, is defined by

$$P_2(2\omega) = \epsilon_0 \chi^{(2)}(-2\omega, \omega, \omega) E(\omega) E(\omega) \quad (7.12)$$

and

$$P_2(0) = 2\epsilon_0 \chi^{(2)}(0, \omega, -\omega) E(\omega) E^*(\omega) \quad (7.13)$$

Where $\chi^{(2)}(-2\omega, \omega, \omega)$ and $\chi^{(2)}(0, \omega, -\omega)$ may both be obtained by putting $\omega_1 = \omega$ and $\omega_2 = +\omega$ and $-\omega$, respectively in the general expression :

$$\chi^{(2)}(-\omega_1 - \omega_2, \omega_1, \omega_2) = -N\lambda \frac{e^3}{m^2} F(\omega_1) F(\omega_2) F(\omega_1 + \omega_2) \quad (7.14)$$

The susceptibility $\chi^{(2)}(-(\omega_1 + \omega_2), \omega_1, \omega_2)$ describes the generation of the polarization component at $\omega_1 + \omega_2$ which is proportional to the product $E(\omega_1)E(\omega_2)$. Similarly we may define the third order susceptibility tensor $\chi^{(3)}(-(\omega_1 + \omega_2 + \omega_3), \omega_1, \omega_2, \omega_3)$ which describes the creation of a polarization $P(\omega_1 + \omega_2 + \omega_3)$ that is proportional to $E(\omega_1)E(\omega_2)E(\omega_3)$.

$$\chi^{(3)}(-\omega_1, -\omega_2, -\omega_3, \omega_1, \omega_2, \omega_3) = \frac{Ne^4}{m^3} \left[\eta + \frac{4}{3} \lambda^2 \{ F(\omega_1 + \omega_2) + F(\omega_2 + \omega_3) + F(\omega_3 + \omega_1) \} \right] \times F(\omega_1) F(\omega_2) F(\omega_3) F(\omega_1 + \omega_2 + \omega_3) \quad (7.15)$$

Note that the susceptibility tensors are properties of the parameter in the potential i.e. properties of the medium. They can be defined in a completely general way.

5. The general expression for $\chi^{(2)}$ suggests that it is proportional to $(\chi^{(1)})^3$. Indeed, in the early days of nonlinear optics Miller [R.C. Miller, Appl. Phys. Lett 5, 17(1964)] found that

$$\chi_{ijk}^{(2)}(\Omega, \omega_1, \omega_2) = \Delta \chi_{ii}^{(1)}(\Omega) \chi_{jj}^{(1)}(\omega_1) \chi_{kk}^{(1)}(\omega_2) \quad (7.16)$$

where Δ was found to be nearly independent of the material. Similar relations were also tried for the third order response but were not so successful. We shall return to this discussion in the lecture on nonlinear optical materials.

RECAP

In this lecture we have learned about the

- Anharmonic oscillator model is used to obtain second and third order susceptibilities.
- Millers rule for second order response is discussed.
- Third order response may arise from lowest order anharmonic term as well as the next order one.