Quantum theory of light-matter interaction: Fundamentals

Lecture 3
Nonlinear dipole oscillator, classical nonlinear optics

Péter Földi

University of Szeged, Dept. of Theoretical Physics, 2014

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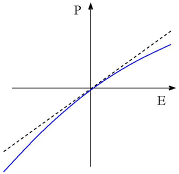
Reminder

For weak external fields, we have $\mathbf{P}(\omega) = \epsilon_0 \chi(\omega) \mathbf{E}(\omega)$, where (for homogeneous media) the susceptibility χ is a complex valued function of the frequency. The Lorentz model (assuming harmonically bound classical electrons) results in $\chi(\omega) \propto 1/\left(\omega_0^2 - \omega^2 - i\gamma\omega\right)$, where γ describes the damping. Note that this expression leads to a causal **P–E** relation.

Nonlinearities

However, when the external field is strong enough (e.g., in solids: comparable to the interatomic electric fields ranging between 10^5–10^8 V/m), nonlinear effects inevitably appear. The figure illustrates the typical situation in this parameter range. During the current lecture, we consider a nonlinear extension of the Lorentz model in order to gain a basic understanding of nonlinear light-matter interactions.



Experimental relevance of nonlinear effects

Generation of difference and sum frequencies

Considering two-color excitation (ω and ω'), nonlinear effects lead to polarization response oscillating also at the difference $\omega - \omega'$ and the sum $\omega + \omega'$ of the exciting frequencies.

High harmonics generation (HHG)*

A strong enough excitation corresponding to a single carrier frequency ω can lead to measurable harmonics $n \times \omega$ up to orders of n around 50.

Ultrashort pulses*

Having multiple harmonics with appropriate phase (frequency domain) can lead to extremely short (attosecond) laser pulses (time domain).

* Theoretical description is beyond the perturbative methods presented here.

Basic concepts

Similarly to the Lorentz model, we consider a single active electron in the atom as a classical point charge $e_0 (< 0)$ of mass m. A damping force should also be included on physical grounds.
The exciting field is assumed to be linearly polarized, monochromatic: $\mathbf{E}(z,t) = E(z,t)\hat{\mathbf{x}}$, with

$$E(z,t) = E_0 \cos(\omega t - kz) = \frac{1}{2} E_0 e^{-i(\omega t - kz)} + c.c..$$

That is, the wave propagates along the z axis, while it is polarized in the x direction. (Note that x is also used to label the deviation of the electron from the equilibrium position with the center of charge at the nucleus. Generally $x = x(z,t)$, but the spatial dependence will be omitted at this point.)

Dynamical equation

The previous assumptions lead to

$$\ddot{x}(t) + \gamma \dot{x}(t) + \omega_0^2 x(t) + a x^2(t) + b x^3(t) + \ldots = \frac{e_0}{m} E_0 \cos(\omega t), \quad \text{(NlinOsc)}$$

where the most important nonlinear terms proportional to a and b explicitly appear.

Notes

- The quadratic term in (NlinOsc) is absent for e.g., isolated atoms (for symmetry reasons).
- In the following we shall not consider higher order terms than the cubic one proportional to b above.

Series expansion

In order to see the role of various orders of approximation, the solution of Eq. (NlinOsc) is usually assumed to have a form of a power series:

$$x(t) = \sum_{n=1}^{\infty} x^{(n)}(t),$$

where, according to the previous lecture,

$$x^{(1)}(t) = \frac{e_0}{m} \frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} \frac{1}{2} E_\omega e^{-i\omega t} + c.c.$$

Higher order terms (corresponding to solutions with increasing accuracy) are obtained by successive approximation. E.g., assuming quadratic nonlinearity,

$$\sum_{n=1}^{N+1} \frac{d^2}{dt^2} x^{(n)} + \gamma x^{(n)} + \omega_0^2 x^{(n)} + a \left[\sum_{n=1}^{N} x^{(n)} \right]^2 = \frac{e_0}{2m} E_0 e^{-i\omega t}. \qquad \text{(Sum)}$$

Details

Let us investigate the simplest case of Eq. (Sum) with $N = 1$. Note that – by construction – the terms linear in the first order solution $x^{(1)}$ cancel the driving field. That is, we have:

$$\frac{d^2}{dt^2}x^{(2)} + \gamma x^{(2)} + \omega_0^2 x^{(2)} = -a\left[x^{(1)}\right]^2.$$

Let us observe that $[x^{(1)}(t)]^2$ can be written as a sum of a constant (dc) term and an additional one oscillating with 2ω. Therefore the form

$$x^{(2)}(t) = \frac{1}{2}\left[x_{dc}^{(2)} + x_{2\omega}^{(2)}e^{-2i\omega t} + c.c.\right]$$

is plausible. By substituting this expression back to the equation on the top of the slide and equating terms with the same time dependencies, we can calculate the coefficients $x_{dc}^{(2)}$ and $x_{2\omega}^{(2)}$, i.e., we can obtain a solution up to second order.

Second order solution for quadratic nonlinearity

$$x_{dc}^{(2)} = -\frac{a}{2\omega_0^2}\left|x^{(1)}(0)\right|^2, \quad x_{2\omega}^{(2)} = -\frac{a}{2}\left(x^{(1)}(0)\right)^2 \frac{1}{\omega_0^2 - (2\omega)^2 - i\gamma 2\omega}. \tag{dc2w}$$

These coefficients provide the leading order nonlinear correction when there is no inversion symmetry and quadratic nonlinearities appear. Note that around resonance, both $x_{dc}^{(2)}$ and $x_{2\omega}^{(2)}$ are divided by essentially ω_0^2, i.e., these corrections are usually small.

Let us recall, that the appearance of the second harmonics in the solution above was due to the fact that this frequency component was present already in the term $\left[x^{(1)}(t)\right]^2$. When the leading order nonlinearity is cubic, $x^{(1)}(t)$ has to be raised to third power (instead of the second), and a term oscillating with 3ω also appear.

Two-color excitation

Let us consider

$$E(z,t) = E_{10}\cos(\omega_1 t + kz) + E_{20}\cos(\omega_2 t + kz). \tag{2color}$$

Following the same route as before, we easily see that the polarization

$$P(z,t) = \mathcal{N} e_0 x(z,t)$$

will contain terms with spatiotemporal dependences of $\cos[(k_1 z - \omega_1 t)]$, $\cos[(k_2 z - \omega_2 t)]$, and $\cos[(k_\pm z - \omega_\pm t)]$, where $\omega_\pm = \omega_1 \pm \omega_2$, and $k_\pm$ are the corresponding wave numbers.

Assuming quadratic nonlinearity, $x_+^{(2)}$ and $x_-^{(2)}$ can be obtained as a straightforward generalization of Eq. (dc2w).

Coupled mode equations

As a simple generalization, let us assume that the amplitudes in Eq. (2color) are slowly varying functions of space and time $E_1 = E_1(z,t)$ and $E_2 = E_2(z,t)$. According to the previous discussion, two additional field modes also arise as a consequence of the nonlinearity:

$$E_{\pm}(z,t) = E_{0\pm}(z,t)\cos(\omega_{\pm} t + kz).$$

According to the first lecture, the slowly varying amplitude equations in steady state read:

$$\frac{\partial}{\partial z} E_{i0}(z,t) = -\frac{k}{2\epsilon_0}\,\mathrm{Im}\,P_{i0}(z,t), \qquad \text{(Amp)}$$

where $P_{i0}(z,t)$ denote the (complex valued) slowly varying amplitude of the corresponding polarization, $i = 1, 2, +, -$.

Coupled mode equations II

In general, different modes are not independent (in fact, the presence of modes 1 and 2 in the nonlinear medium is the origin of the appearance of modes + and -). Although it is not visible explicitly in Eq. (Amp), this fact appears as the dependence of the polarization corresponding to a given mode on the amplitude of different modes as well. (Recall again the case of second harmonic, sum or difference frequency generation.) The resulting complex system of coupled equations is usually relatively difficult to solve.

However, in order get an insight into the physical mechanisms, we can rely on simple approximations. Let us concentrate on difference frequency generation, and assume on physical grounds that the weak mode '-' does not act back on the considerably more intense modes 1 and 2. This (and arguments to be prested a little later) allows us to use the linear theory for P_{10} and P_{20}, and consider the consequences of second order nonlinearity only for P_{-0}.

Coupled mode equations III

Using the assumptions summarized on the previous slide, it can be shown [1] that in our simple collinear model (all fields propagate along the z axis), the intensity of the weak difference frequency signal is given by

$$I_{-}(L) \propto I_1 I_2 \frac{sin^2(\Delta k L/2)}{(\Delta k L/2)^2}, \qquad \text{(Iminus)}$$

where L is the length of the nonlinear medium (typically a crystal), and $\Delta k = k_1 - k_2 - k_{-}$. The maximum of the function above is reached for $\Delta k = 0$, which is an appearance of momentum conservation. However, for collinear propagation, Δk can only vanish for dispersionless media.

Phase matching

If $\Delta k \neq 0$ in Eq. (Iminus), the output intensity $I_{-}(L)$ will be an oscillating function of ΔkL : maxima and minima appear according to constructive and destructive interferences at the output point L.
Maxima of (Iminus) appear periodically at

$$\Delta kL = m\pi,$$

with m being integer. When this *phase matching condition* is fulfilled for e.g., difference frequency generation, additional nonlinear effects (like sum frequency generation, appearance of harmonics for any of the two exciting frequencies) are suppressed. The reason is that the conditions $(k_1 - k_2 - k_{-})L = m\pi$ and, e.g., $(k_1 - k_2 - k_{+})L = m\pi$ are incompatible, they cannot be satisfied at the same time. That is, phase matching is usually possible only for a given nonlinear process.

Note that in 3D, the vectorial version of the phase matching condition has to be fulfilled, e.g., by using birefringent media.

A reminder on causality

The polarization **P** is usually generated by an external electric field **E** in a medium with polarizable atoms, and it is also a source of a secondary electric field. In both cases, causality requires that even in the presence of memory effects, **P** at a certain time instant t and spatial point $\mathbf{r}$ cannot depend on $\mathbf{E}(\mathbf{r}', t')$ if $t' > t$. In other words, the most general **P–E** relation in a linear medium is the following:

$$P_i(\mathbf{r}, t) = \epsilon_0 \int d^3\mathbf{r}' \int_{-\infty}^{t} dt' \chi_{ij}(\mathbf{r} - \mathbf{r}', t - t') E_j(\mathbf{r}', t'). \qquad \text{(Causality)}$$

In frequency-wavenumber domain (after 4D Fourier transform) the convolutions above become a local product, i.e., we have

$$P_i(\mathbf{k}, \omega) = \chi_{ij}(\mathbf{k}, \omega) E_j(\mathbf{k}, \omega).$$

Note that any physically relevant expression for $\chi_{ij}(\mathbf{k}, \omega)$ must respect (Causality). The oscillator model of Lecture 2 can be seen to be causal.

Nonlinear susceptibility

The nonlinear version of Eq. (Causality) to second order nonlinearity can be written as

$$\frac{P_i(\mathbf{r},t)}{\epsilon_0} = \text{linear contribution}$$

$$+ \iint \int_{-\infty}^{t} \int_{-\infty}^{t} d^3\mathbf{r}' d^3\mathbf{r}'' dt' dt'' \chi^{(2)}_{ijk}(\mathbf{r}-\mathbf{r}', \mathbf{r}-\mathbf{r}'', t-t', t-t'') E_j(\mathbf{r}', t') E_k(\mathbf{r}'', t'').$$

Generalization to higher orders is straightforward.

In frequency domain (let us omit spatial/wavenumber dependence for now) $\mathbf{P}(\omega)$ is nonzero only if the nonlinear effect we consider allows the appearance of the frequency component ω. E.g., for second order nonlinerity with excitation at ω_1 and ω_2, nonlinear processes induce $\mathbf{P}(\omega_1 - \omega_2)$ and $\mathbf{P}(\omega_1 + \omega_2)$. For sum frequency generation, e.g., we have

$$P_i(\omega_1 + \omega_2) = \chi^{(2)}_{ijk}(\omega_1 + \omega_2) E_j(\omega_1) E_k(\omega_2).$$

Remarks

- When the exciting field is essentially monochromatic (although strong) one usually writes:

$$P_i = \epsilon_0 \sum_j \chi_{ij}^{(1)} E_j + \epsilon_0 \sum_{j,k} \chi_{ijk}^{(2)} E_j E_k + \epsilon_0 \sum_{j,k,l} \chi_{ijkl}^{(3)} E_j E_k E_l + \ldots .$$

- We have not mentioned so far the possible appearance of combination tones: For higher order processes, assuming excitation at frequencies ω_1 and ω_2, the polarization that arise will contain frequency components $n\omega_1 + m\omega_2$, with n, m being integer.

Outlook

Nonlinear optical phenomena cover a wide, experimentally important area. During the current lecture we introduced a transparent, classical model that helped us to see the basic concepts in this field. Obviously, sophisticated quantum mechanical models can provide a more realistic description, but this is beyond the scope of the current course.

Special nonlinear effects that are of experimental importance (like 3 and 4 wave mixing together with the corresponding phase matching conditions) will be discussed in the next lecture.

Questions

1. Summarize the assumptions of the Lorentz model.
2. In what way have we generalized the Lorentz model in this lecture?
3. Considering strong, monochromatic excitation, and quadratic nonlinearity, what frequency components does the induced polarization have?
4. Considering strong, monochromatic excitation, and cubic nonlinearity, what frequency components does the induced polarization have?
5. Explain the physical mechanism that can couple different modes of electromagnetic radiation traversing a nonlinear crystal.
6. What is phase matching?

Questions (continued)

7. Can the phase matching conditions be fulfilled (in general) for different nonlinear processes?
8. In what sense does the notion of causality appear in **P**– **E** relations?
9. Explain, why the linear relation between **P** and **E** is local in the variables **k** and ω.
10. What is the physical role of nonlinear susceptibility?

Reference

1. P. Meystre and M. Sargent, *Elements of Quantum Optics*, Spinger (Berlin, Heidelberg) (2007).