TFY4200 - Problem set 7 (6)
A self study on non-linear optics

Svein Åmdal

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Non-linearity of scalar fields

Previously, we have taken the dielectric response to an applied E-field (generally from a EM-wave) to be linear, meaning that $D = \epsilon_0 \epsilon E$ holds. Now, we focus on the dielectric polarization P and assume it might be non-linear, i.e.

$$P(t) = \epsilon_0 \sum_{n=1}^{\infty} \chi^{(n)} E^n(t) = \sum_{n=1}^{\infty} P^{(n)}(t). \tag{1}$$

$\{\chi^{(n>1)}\}$ is called the *higher order non-linear optical suceptibilites.*

Second-order non-linear optical processe

For example, if a laser whose field is given by

$$E_{\text{inc}}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_1^* e^{i\omega_1 t} + E_2^* e^{i\omega_2 t} \tag{2}$$

is incident on a crystal with $\chi^{(2)} \neq 0$, we calculate

$$\begin{aligned} P^{(2)}(t) &= \epsilon_0 \chi^{(2)} E^2(t) \\ &= \epsilon_0 \chi^{(2)} \Big[E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + E_1^{*2} e^{2i\omega_1 t} + E_2^{*2} e^{2i\omega_2 t} \\ &+ 2E_1 E_2 e^{-i(\omega_1+\omega_2)t} + 2E_1^* E_2^* e^{i(\omega_1+\omega_2)t} + 2E_1 E_2^* e^{-i(\omega_1-\omega_2)t} \\ &+ 2E_1^* E_2 e^{i(\omega_1-\omega_2)t} + 2E_1 E_1^* + 2E_2 E_2^* \Big] \\ &:= \sum_n P(\omega_n) e^{-i\omega_n t}. \end{aligned} \tag{3}$$

We now identify

$$P(2\omega_1) = \epsilon_0 \chi^{(2)} E_1^2 \tag{4}$$

$$P(2\omega_2) = \epsilon_0 \chi^{(2)} E_2^2, \tag{5}$$

as *second-harmonic generation* (SHG) of polarization response. These contributions to the polarization response has twice the incoming frequency, and they stem from interaction between only a single frequency (either ω_1 or ω_2). Thus, SHG is also apparent if the incoming wave consists of only a single frequency component.

Further, we identify

$$P(\omega_1 + \omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2, \tag{6}$$

as *sum-frequency generation* (SFG) of polarization response, and

$$P(\omega_1 - \omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2^*, \tag{7}$$

as *difference-frequency generation* (DFG) the polarization response. Note that eqs. (4) to (7) all have corresponding negative frequency components, which we ommit.

Finally, we may identify

$$P(0) = 2\epsilon_0 \chi^{(2)} \Big[E_1 E_1^* + E_2 E_2^* \Big], \tag{8}$$

as the DC components of P, often called *optical rectification*, a quantity that is also present with only a single incoming frequency.

If the crystal has few avaliable states with the bandgap energies $\hbar\omega_{\{\text{inc}\}}$, almost all the power of the incident wave is immediately radiated with SHG frequency $2\omega_{\text{inc}}$, or with one of the sum- or difference-generated frequencies. One application of this is to produce tunable radiation. If the input frequencies are produced by one fixed-ω and one tunable-ω laser, the resulting radiation is tunable, and may be in a different frequency regime than the tunable laser.

Third-order non-linear optical processes

For simplicity, let the incoming field be monochromatic, given by

$$E_{\text{inc}}(t) = E \cos(\omega t). \tag{9}$$

Using a mathematical cosine identity, we find

$$\begin{aligned} P^{(3)}(t) &= \epsilon_0 \chi^{(3)} E_{\text{inc}}^3(t) \\ &= \frac{1}{4} \epsilon_0 \chi^{(3)} E^3(t) \Big[\cos(3\omega t) + 3\cos(\omega t) \Big]. \end{aligned} \tag{10}$$

From eq. (1), we have that the third-order polarization response is

$$P^{(3)}(t) = \epsilon_0 \chi^{(3)} E^3(t). \tag{11}$$

Similarly to the second-harmonic generation of polarization, we now get a *third-harmonic generation* of polarization, namely the 3ω-term.

For the more general case of a tri-chromatic incoming wave, we get generation of all possible frequencies that are sums and/or differences between the different incoming frequencies.

The processes discussed so far are all *parametric*, meaning that they do not alter the quantum state of the crystal (in any meaningful way, at least). If the final state of the crystal has a sustantial number of excited electrons compared to the initial state, the process is *non-parametric*, and must be described by a complex χ. In a non-parametric process, the photon energy may not be conserved, as it may become absorbed in the medium. This is similar to the familiar case of a complex refractive index being a measure of wave dissipation in the medium.

Non-linearity of vector fields (formal treatment)

We begin by writing out the electric field as a sum of frequency components (both positive and negative), i.e.

$$\boldsymbol{E}(\boldsymbol{r},t) = \sum_n \boldsymbol{E}_n(\boldsymbol{r},t) = \sum_n \boldsymbol{A}_n e^{i(\boldsymbol{k}_n\cdot\boldsymbol{r}-\omega_n t)}, \tag{12}$$

where

$$\boldsymbol{A}_n = \boldsymbol{A}(\omega_n) = \boldsymbol{A}(-\omega_n) = |\boldsymbol{E}_n(\boldsymbol{r})| = \frac{1}{2}E. \tag{13}$$

(The factor $1/2$ comes from counting both positive and negative ω in the sum.) Now, the second order susceptibility tensor $\chi^{(2)}_{ijk}$ is defined in terms of the cartesian components i, j and k of $\boldsymbol{E}$ by the equation

$$P_i(q_{mn} = \epsilon_0 \sum_j \sum_k \sum_{m,n|q_{mn}=\text{const.}} \chi^{(2)}_{ijk}(q_{mn};\omega_m,\omega_n)E_j(\omega_m)E_k(\omega_n), \tag{14}$$

where q_{mn} is shorthand for $\omega_m+\omega_n$, and it is assumed to be fixed in the sum over m and n.

By letting $m,n\in\{1,2\}$ and $q=\omega_1+\omega_2$, one automatically recieves sum-frequency generation of a response with frequency $\omega_1+\omega_2$ exactly like before.

By letting ω_1 be given, and letting $q=2\omega_1$, one automatically recieves second-harmonic generation of response polarization, also like before.

Precisely analoguosly, the third order susceptibility tensor $\chi^{(3)}_{ijkl}$ is defined by

$$P_i(q_{mno}) = \epsilon_0 \sum_{j,k,l} \sum_{m,n,o|q_{mno}=\text{const.}} \chi^{(3)}_{ijkl}(q_{mno};\omega_m,\omega_m,\omega_o)E_j(\omega_m)E_k(\omega_n)E_l(\omega_o), \tag{15}$$

where $q_{mno}=\omega_m+\omega_n+\omega_o$. In both cases we may perform the summation over m,n (and o), and collect the result in a multiplicative *degeneracy factor*, D.

Anharmonic oscillator

The classical harmonic oscillator for a position-like parameter x only has terms up to second order in x. The first order term may be removed by a coordinate transformation, and the constant term may be removed by carefully selecting the zero-level of the harmonic potential. Let's now introduce a x^3-anharmonic term. Thus the oscillator potential becomes

$$U(x) = \frac{1}{2}m\omega_0^2 x^2 + \frac{1}{3}max^3, \tag{16}$$

for some choices of parameters m, ω_0 and a. The anharmonic force from displacement in this potential becomes of second order in x, as

$$F = -\frac{\partial}{\partial x}U(x) = -m\omega_0^2 x - max^2. \tag{17}$$

Inserting this "external" force into the classical equation of motion for an electron (charge e) with a damping factor γ, and an external, di-chromatic E-field given by eq. (2), we solve the modified equation of motion,

$$\ddot{x} + 2\gamma\dot{x} + \omega_0^2 x + ax^2 = -\lambda\frac{eE(t)}{m}, \tag{18}$$

in terms of a perturbation expansion for small x in the now introduced expansion parameter λ, i.e.

$$x = \sum_n \lambda^n x^{(n)}. \tag{19}$$

Collecting the terms of equal order in λ, we find that the 1^{st} order terms correspond to the unperturbed equation of motion. Thus

$$\begin{aligned} x^{(1)}(t) = &-\frac{e}{m}\frac{E_1}{\omega_0^2 - \omega_1^2 - 2\gamma i\omega_1}\exp\{-i\omega_1 t\} \\ &-\frac{e}{m}\frac{E_2}{\omega_0^2 - \omega_2^2 - 2\gamma i\omega_2}\exp\{-i\omega_2 t\} \\ &-\frac{e}{m}\frac{E_1}{\omega_0^2 - \omega_1^2 + 2\gamma i\omega_1}\exp\{i\omega_1 t\} \\ &-\frac{e}{m}\frac{E_2}{\omega_0^2 - \omega_2^2 + 2\gamma i\omega_2}\exp\{i\omega_2 t\}. \end{aligned} \tag{20}$$

The 2^{nd} order terms collect to form the equation

$$\ddot{x}^{(2)} + 2\gamma\dot{x}^{(2)} + \omega_0^2 x^{(2)} + a(x^{(1)})^2 = 0. \tag{21}$$

Inserting eq. (20) into eq. (21), we obtain an expression for $x^{(2)}$. If one defines

$$D(\omega_j) := \omega_0^2 - \omega_j^2 - 2\gamma i\omega_j, \tag{22}$$

one may write down the frequency components of $x^{(2)}$ instead.

Given the known linear polarization

$$P^{(1)}(\omega) = \epsilon_0 \chi^{(1)}(\omega) E(\omega) = -Nex^{(1)}(\omega), \tag{23}$$

we may calculate

$$\chi^{(1)}(\omega_j) = \frac{Ne^2/m}{\epsilon_0 D(\omega_j)} \tag{24}$$

based on eqs. (20) to (23). Defining non-linear polarization as

$$P^{(2)}(q_{12}) = 2\epsilon_0 \chi^{(2)}(q_{12}; \omega_1, \omega_2) E(\omega_1) E(\omega_2) = -Nex^{(2)}(q_{12}), \tag{25}$$

still using the shorthand $q_{ij} = \omega_i + \omega_j$, we analogously find

$$\begin{aligned} \chi^{(2)}(q_{12}; \omega_1, \omega_2) &= \frac{Nae^3/m^2}{\epsilon_0 D(q_{12}) D(\omega_1) D(\omega_2)} && (26) \\ &= \frac{\epsilon_0^2 ma}{N^2 e^3} \chi^{(1)}(q_{12}) \chi^{(1)}(\omega_1) \chi^{(1)}(\omega_2), && (27) \end{aligned}$$

where we in the last equality have used eq. (24). This corresponds to sum-frequency generation. A special case of this is when $\omega_1 = \omega_2 = \omega$. Then

$$\chi^{(2)}(2\omega; \omega, \omega) = \frac{\epsilon_0^2 ma}{N^2 e^3} \chi^{(1)}(2\omega) [\chi^{(1)}(\omega)]^2, \tag{28}$$

and the polarization resulting from this is akin to second-harmonic generation.

We may also have $\omega_2 \to -\omega_2$. Then,

$$\chi^{(2)}(q_{1,-2}; \omega_1, -\omega_2) = \frac{\epsilon_0^2 ma}{N^2 e^3} \chi^{(1)}(q_{1,-2}) \chi^{(1)}(\omega_1) \chi^{(1)}(-\omega_2), \tag{29}$$

and we have arrived at difference-frequency generation of polarization.

A third special case is when $\omega_2 = -\omega_1 := -\omega$. Then

$$\chi^{(2)}(0, \omega, -\omega) = \frac{\epsilon_0^2 ma}{N^2 e^3} \chi^{(1)}(0) \chi^{(1)}(\omega) \chi^{(1)}(-\omega) \tag{30}$$

represents optical rectification.

A similar analysis is performed for the tensor form of $\chi^{(2)}$ to obtain

$$\begin{aligned} \chi^{(2)}_{ijkl}(q_{mno}, \omega_m, \omega_n, \omega_o) = &\frac{bm\epsilon_0^3}{3N^3 e^4} \chi^{(1)}(q_{mno}) \chi^{(1)}(\omega_m) \chi^{(1)}(\omega_n) \chi^{(1)}(\omega_o) \\ &\times \left[\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} \right], \end{aligned} \tag{31}$$

where χ_{ijkl} means the susceptibility in direction i from $\boldsymbol{E}$ with components in directions j, k and l.