Solution to Problem Set 2
Nonlinear Optics (NLO)

Due: 29. April 2015

1) Typical electric field strengths in nonlinear optics, typical nonlinear susceptibilities

For an order-of-magnitude estimation of the nonlinear susceptibilities $\chi^{(2)}$ and $\chi^{(3)}$, let us consider a simple model of a Hydrogen atom, in which the electron is bound to the nucleus by an electric field with the radial component that corresponds to a characteristic atomic field strength

$$E_{\text{at}} = \frac{e}{\epsilon_0 4\pi a_0^2}, \tag{1.1}$$

where $e = 1.60\times10^{-19}\,\text{C}$ is the elementary charge, $\epsilon_0 = 8.85\times10^{-12}\,\text{F/m}$ is the vacuum permittivity and $a_0 = 4\pi\epsilon_0 \hbar^2/m_e e^2 = 0.053\,\text{nm}$ is the Bohr radius, i.e. the expectation value of the distance between the nucleus and the electron. The quantity $\hbar = 6.626\times10^{-34}\,\text{Js}$ denotes the reduced Planck constant, and $m_e = 9.1\times10^{-31}\,\text{kg}$ is the electron mass. Externally applied electric fields lead to a displacement of the electron with respect to the core and hence to an electric dipole moment, i.e., a non-zero electric polarization P. Usually external fields (E_{ext}) are much smaller than E_{at} and the dependence between E_{ext} and P can be approximated by the linear relationship $P = \epsilon_0\chi^{(1)}E_{\text{ext}}$. For most solid-state materials, $\chi^{(1)}$ is in the order of unity.

A common argument [1] says that the nonlinear contributions to the polarization $P_{\text{NL}}^{(m)} = \epsilon_0\chi^{(m)}E^m$ ($m > 1$) become comparable to the linear contribution $P_{\text{L}} = \epsilon_0\chi^{(1)}E$ when the applied electric field E_{ext} is of the order of E_{at}.

1. Calculate the characteristic atomic field strength E_{at}. Then, set $\chi^{(1)} = 1$ and assume for the radial component of the polarization that $P_{\text{L}} = P_{\text{NL}}^{(m)}$ for E_{ext} = E_{at}. Use these relations to estimate the order of magnitude of $\chi^{(2)}$ and $\chi^{(3)}$.

A typical femtosecond laser system produces pulses with a repetition rate of 80 MHz, a pulse duration of 100 fs, and an average power of 1 W at a wavelength of 800 nm. Imagine that the beam is focused on a spot having diameter equal to the wavelength.

2. What is the optical power averaged on one pulse, assuming that the pulse has rectangular shape?

3. What is the maximum electric field strength in the focus assuming that the field is uniform within the given diameter? Compare the magnitude of this field with E_{at}.

[1] Boyd, R.W., *Nonlinear Optics* (San Diego: Academic Press, 2003).

Solution:

1.

$$E_{\text{at}} = 5.13\times 10^{11}\,\frac{\text{V}}{\text{m}}$$

$$\chi^{(2)} = \frac{1}{E_{\text{at}}} = 1.95\times 10^{-12}\,\frac{\text{m}}{\text{V}}$$

$$\chi^{(3)} = \frac{1}{E_{\text{at}}^2} = 3.81\times 10^{-24}\,\frac{\text{m}^2}{\text{V}^2}$$

2.

$$P_{\text{avg}} = 1\text{W}$$

$$P_{\text{pulse}} = \frac{12.5\text{ns}}{100\text{fs}}\cdot P_{\text{avg}} = 125\text{kW}$$

3.

$$I_{\text{pulse}} = \frac{P_{\text{pulse}}}{\pi\left(0.5\mu\text{m}\right)^2} = 2.5\times 10^{17}\,\frac{\text{W}}{\text{m}^2}$$

$$E_{\text{pulse}} = \sqrt{\frac{2I_{\text{pulse}}}{c\epsilon_0}} = 1.4\times 10^{10}\,\frac{\text{V}}{\text{m}}$$

where c is the speed of light in vacuum. This field is about a factor of 40 less than E_{at}, however this can be resolved with the sensitivity of today's equipment.

2) Third-order nonlinear polarization

Consider a linearly polarized plane wave $\mathbf{E}(z,t) = E(z,t)\mathbf{e}_x$ propagating in z-direction in a homogeneous medium, in which third-order nonlinear effects dominate over second and higher-order contributions, $\chi^{(3)} \neq 0$, $\chi^{(m)} = 0$ for m = 2 or m > 3. Assuming an instantaneous response of the polarization P_{NL} to the applied electric field, we can express P_{NL} as

$$P_{\text{NL}}(z,t) = P^{(3)}(z,t)\mathbf{e}_x\text{, with } P^{(3)}(z,t) = \varepsilon_0\chi^{(3)}E(z,t)^3\,. \tag{2.1}$$

Calculate $P^{(3)}(z,t)$ considering a field $E\left(z,t\right)$ composed of two distinct frequency components ω_1 and ω_2 with their complex amplitudes $\underline{E}_1$ and $\underline{E}_2$

$$E(z,t) = \text{Re}\left\{\underline{E}_1 e^{\text{j}(\omega_1 t - k_1 z)} + \underline{E}_2 e^{\text{j}(\omega_2 t - k_2 z)}\right\}\;. \tag{2.2}$$

Group the resulting terms with appropriate degeneracy factors according to their frequency and assign them to the following effects:

- Third-Harmonic Generation (THG)
- Third-order Sum-Frequency Generation (TSFG)
- Self-Phase Modulation (SPM)
- Cross-Phase Modulation (XPM)
- (Degenerate) Four-Wave Mixing (FWM)

Solution:

First, we reformulate (2.2) in a complex notation,

$$E(z,t)=\frac{1}{2}\left\{E_1 e^{\mathrm{j}(\omega_1 t-k_1 z)}+E_2 e^{\mathrm{j}(\omega_2 t-k_2 z)}+c.c.\right\}.$$

As we will calculate the third power of four factors, we expect a total number of 4^3=64 terms, which can be expressed as

$$\begin{aligned}
P^{(3)}(z,t)=\frac{1}{8}\varepsilon_0\chi^{(3)}\cdot\\
&[E_1^3 e^{j(3\omega_1 t-3k_1 z)}+E_2^3 e^{j(3\omega_2 t-3k_2 z)}+c.c.\\
&+3E_1^2 E_2 e^{j((2\omega_1+\omega_2)t-(2k_1+k_2)z)}+3E_2^2 E_1 e^{j((\omega_1+2\omega_2)t-(k_1+2k_2)z)}+c.c.\\
&+3\left|E_1\right|^2 E_1 e^{j(\omega_1 t-k_1 z)}+3\left|E_2\right|^2 E_2 e^{j(\omega_2 t-k_2 z)}+c.c.\\
&+6\left|E_1\right|^2 E_2 e^{j(\omega_2 t-k_2 z)}+6\left|E_2\right|^2 E_1 e^{j(\omega_1 t-k_1 z)}+c.c.\\
&+3E_1^2 E_2^* e^{j((2\omega_1-\omega_2)t-(2k_1-k_2)z)}+3E_2^2 E_1^* e^{j((-\omega_1+2\omega_2)t-(-k_1+2k_2)z)}+c.c.].
\end{aligned}$$

The resulting terms can be grouped by frequencies:

Frequency	Degeneracy	Name
$3\omega_1$, $3\omega_2$	1	THG – third harmonic generation
$2\omega_1+\omega_2$, $2\omega_2+\omega_1$	3	TSFG – third order sum-frequency generation
$2\omega_1-\omega_2$, $2\omega_2-\omega_1$	3	FWM – (degenerate) four-wave mixing
ω_1 from $(\omega_2;-\omega_2;\omega_1)$, ω_2 from $(\omega_1;-\omega_1;\omega_2)$	6	XPM – cross-phase modulation
ω_1 from $(\omega_1;\omega_1;-\omega_1)$, ω_2 from $(\omega_2;\omega_2;-\omega_2)$	3	SPM – self-phase modulation

For three frequencies the general rule is that all frequencies $\omega=\left[\pm\omega_l\pm\omega_m\pm\omega_n\right]$ for $l,m,n\in\{1,2,3\}$ are generated. The degeneracy factor is given by the number of possible permutations of those frequencies. For each 'negative' frequency the complex conjugate of the respective field component has to be taken, e.g. $2\omega_1-\omega_2\rightarrow E_1E_1E_2^*$.

Note: Here TSFG and DFWM are classified as different processes but FWM embraces also TSFG and, in fact, all third order nonlinear processes can be considered as FWM processes. Different authors could name in slightly different way some of the corresponding third order nonlinear processes.

Questions and Comments:

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