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# "Introduction to Second Order Nonlinear Processes"



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Introduction to second order nonlinear processes.

We will begin by describing the first nonlinear optical experiment using a laser source and heuristically developping an explanation for the observed phenomena. We will then go on to present a more complete formalism describing general three wave interactions in quadratically nonlinear materials.

The first nonlinear optical experiment using lasers as the source was that of Franken et al in 1961. In this experiment the light from a pulsed ruby laser (λ = .6943 μm) was focussed on a quartz crystal and the generation of second harmonic light (λ= .3471 μm) was observed. This harmonic light is created by a nonlinear polarization proportional to the square of the incident optical field. Furthermore, as the crystal was rotated, with the rotation axis normal to the incident beam, a periodic variation of the second harmonic light power was observed which the authors correctly ascribed to the effect of *phase matching*. This effect, which is of extreme importance in practically all nonlinear experiments, arises from the fact that the nonlinear polarization, which serves as the *source* for the second harmonic radiation, does not have the same phase velocity as the second harmonic radiation itself. This is because, for a plane wave propagating in the z direction, the nonlinear polarization is proportional to $E_\omega^2(z)e^{i2(\omega t-k_\omega z)}$ and has a phase velocity given by $\frac{2\omega}{2k}=\frac{\omega}{k}$ which is simply the phase velocity of the fundamental beam at frequency ω. On the other hand, the harmonic beam will have a phase velocity given by $\frac{2\omega}{k_{2\omega}}$ and since $k_{2\omega}\neq 2k\omega$ the nonlinear polarization and the harmonic wave have different phase velocities. To calculate the effect of this, we consider the simplest possible case, a plane wave at frequency ω incident on a nonlinear material. If the thickness of the nonlinear material is l, the harmonic field at the output will be proportional to

$$E_{2\omega}(l)\propto\int_0^l e^{2i(\omega t-kz)}e^{-ik_{2\omega}(l-z)}dz$$

and, a simple calculation leads to $E_{2\omega}(l)\propto\frac{e^{i\Delta kl}-1}{i\Delta k}$, or, in terms of power,

$P_{2\omega}(l)\propto L^2\operatorname{sin}c^2\left(\frac{\Delta kl}{2}\right)$, where $\Delta k=k_{2\omega}-2k_\omega$, or, $\Delta k=2k_0(n_{2\omega}-n_\omega)$ and $k_0$ is the fundamental vacuum k vector.

We define the coherence length of such as interaction as the length at which the second harmonic will reach it's maximum before diminishing, i. e. $l_c=\frac{\pi}{\Delta k}$

As an example, if we have λ = 1μm, Δn = $10^{-2}$ then $l_c$ is on the order of 50 μm. After this distance the propagating and the newly generated wave will interfere destructively and reduce the second harmonic power. This leads to a very strong limit of conversion efficiency.

There are three main techniques used to attain the phase matching condition; $\Delta k=0$.

These are, in the order of their historical appearance :

-birefringence
- "Cerenkov" phase matching
- quasi-phase-matching

In birefringent phase-matching one takes advantage of the fact that in certain crystals there is an equality of the fundamental and harmonic indices of refraction, for a certain angle of

propagation, when one of the waves has ordinary polariztion and the other, extraordinary polarization.
In "Cerenkov" phase matchng the second harmonic propagates at an angle such that the projection of the second harmonic k vector on the direction of propagation of the fundamental is equal to twice the fundamental wave vector. This type of phase matching always occurs but is only efficient for a small range of angles for which the overlap integral of the fundamental and SH waves takes on reasonable values.
In all commercial nonlinear devices sold today, birefringent phase matching is used. However, due to the order of magnitude higher efficiency posible with QPM, this technique should soon find it's place in practical applications.In this technique one uses periodically inverted domains , i.e. periodic inversion of the sign of the nonlinear coefficient at eac coherence length, to produce a $\pi$ phase shift in the generated wave and thereby ensure constructive, rather than destructive, interference in the following segment. While first-order QPM leads to a reduction of a factor of about 1/2 in the efficiency if the nonlinear coefficients are equal, QPM allows the use of a nonlinear coefficient that is often on the order of 6 times greater than that involved in the birefringnt phase matching scheme (which is not surprising in view of the geometry), resulting in an overall gain of around 20 in the conversion efficiency.
With 100 mw of pump coupled into a waveguide QPM structure, the generation of 15 mW of SHG has been demonstrated which is currently the world record for normalised conversion effiency: 600%/W-cm$^2$ in the non depleted pump approximation.
Since the theoretical limit is approximately twice this factor, some further improvement can be expected. However, to gain order of magnitude improvements new material-microstructure systems appear to be necessary.
After this brief heuristic introduction, we now go on to discuss the source of the nonlinear polarization and it's relation to the symmetry properties of the nonlinear medium.

Nonlinear polarization

We begin by recalling that the linear polarization of a material is given by:
$$p(t) = eNx(t)$$
where e is the charge of the electron, N the interacting electron density, and x the electron deviation from the equilibrium position.The electrons are considered to be oscillators in a potential well described by:
$$V(x) = \frac{m\omega_0^2}{2}x^2 + \frac{m}{3}Dx^3 + ......$$
and the restoring force on the electron is given by:
$$F = -\frac{\partial V(x)}{\partial x} = -(m\omega_0^2 x + mDx^2 + ....)$$
We note that the restoring force is asymmetric: it is larger for a deviation in the + direction than for a deviation in the – direction. This can only occur if the crystal has such an assymetry, i.e. if the crystal is non-centrosymmetric.
We now formally relate the nonlinear polarization to the driving field. We begin with the equation of motion of a driven electron:
$$\frac{d^2x(t)}{dt^2} + \sigma\frac{dx(t)}{dt} + \omega_0^2 x(t) + Dx^2(t) = \frac{eE^{(\omega)}}{2m}(e^{i\omega t} + e^{-i\omega t})$$
We assume a solution of the form:
$$x(t) = \frac{1}{2}\left(q_1 e^{i\omega t} + q_2 e^{2i\omega t} + c.c.\right)$$
substitute this expression into the equation of motion and equate the coefficients of $e^{\pm i\omega t}$ and $e^{\pm 2i\omega t}$. Using the approximation that $|Dq_2| << \left[(\omega_0^2 - \omega^2) + \omega^2\sigma^2\right]$ we obtain:
$$q_1 = \frac{eE^{(\omega)}}{m}\frac{1}{(\omega_0^2 - \omega^2) + i\sigma\omega}$$
For the linear polarization this yields

$$p^{(\omega)}(t) = \frac{eN}{2}(q_1 e^{i\omega t} + c.c.) = \frac{\varepsilon_0}{2}(\chi^{(\omega)} E^{(\omega)} e^{i\omega t} + c.c.)$$

Similarly, we obtain:

$$q_2 = \frac{-De^2 (E^{(\omega)})^2}{2m^2 \left[(\omega_0^2 - \omega^2) + i\sigma\omega\right]^2 (\omega_0^2 - 4\omega^2 + 2i\sigma\omega)}$$

The nonlinear polarization is, therefore:

$$p^{(2\omega)}(t) = -\frac{eN}{2}(q_2 e^{2i\omega t} + c.c.) = \frac{1}{2}\left\{d^{(2\omega)}\left[E^{(2\omega)}\right]^2 e^{2i\omega t} + c.c.\right\}$$

which defines the second order nonlinear optical $d^{(2\omega)}$.We can define the complex nonlinear polarization amplitude as:

$$p^{(2\omega)} = \frac{1}{2}\left[P^{(2\omega)} e^{2i\omega t} + c.c.\right] \text{ and } P^{(2\omega)} = d^{(2\omega)} E^{(\omega)} E^{(\omega)}$$

With these definitions we have:

$$d^{(2\omega)} = \frac{DNe^3}{2m^2\left[(\omega_0^2 - \omega^2) + i\sigma\omega\right]^2 (\omega_0^2 - 4\omega^2 + 2i\sigma\omega)} = \frac{mD\left[\chi^{(\omega)}\right]^2 \chi^{(2\omega)} \varepsilon_0^3}{2N^2 e^3}$$

So far we have been treating this as a scalar problem. In real life the second order nonlinear polarization is related to the electric field by a third rank tensor $d_{ijk}$. This is expressed as:

$$P_x^{(2\omega)} = d_{11}^{(2\omega)} E_x^{(\omega)} E_x^{(\omega)} + d_{22}^{(2\omega)} E_y^{(\omega)} E_y^{(\omega)} + d_{33}^{(2\omega)} E_z^{(\omega)} E_z^{(\omega)} +$$

$$2d_{14}^{(2\omega)} E_z^{(\omega)} E_y^{(\omega)} + 2d_{15}^{(2\omega)} E_x^{(\omega)} E_z^{(\omega)} + 2d_{16}^{(2\omega)} E_x^{(\omega)} E_y^{(\omega)}$$

where we have used the contracted indices introduced in our discussion of the photoelastic effect.

| | | | | | |
|---|---|---|---|---|---|
| 11 | 22 | 33 | 23 | 13 | 12 |
| 1 | 2 | 3 | 4 | 5 | 6 |

It is clear that the existence and the size of the coefficients are a function of the material used. The coefficients have been measured for a large number of crystals and are given in various tabulations using a rather astonishing number of definitions and units. Care must be taken when comparing, and using these coefficients. For the parametric interactions we shall discuss, the "usual" units are pv/m, and, as an example, we can take lithium niobate whose $d_{33}$ coefficient, it's largest is on the order of 40 pm/v, with $d_{31}$ and $d_{22}$ equal to 6 and 3 pm/v respectively.

Surprising as it may seem, people sometimes "forget" that the material must be stable, have low absorbtion, be of good optical quality with reasonable volume, etc. In other words, beware of articles intitled "Monster optical nonlinearity in XXXX ..". Most of the stuff turns out to be useless.

## Parametric Interactions

We begin by adding a nonlinear polarization term to the equation of propagation. In a scalar form this equation becomes:

$$\nabla^2 = \mu\sigma\frac{\partial e}{\partial t} + \mu\varepsilon\frac{\partial^2 e}{\partial t^2} + \mu\frac{\partial^2 p_{nl}(\vec{r},t)}{\partial t^2}$$ where we have taken, for simplicity, $p_{nl}$ parallel to e.

where $e^{\omega 2} = \frac{1}{2}\left[E_2(z) e^{i(\omega_2 t - k_2 z} + c.c.\right]$ and $p_{nl})_i = d_{ijk} E_j E_k$ and we recognize that d is a third rank tensor.

We now consider the general case of parametric interactions; that of three plane waves of different frequencies propagating in the z direction. The example of second harmonic generation is a special case where two of the waves have the same, fundamental, frequency.

$$e^{\omega i} = \frac{1}{2}\left[E_i(z)e^{i(\omega_i t + k_i z)} + c.c.\right]$$ with $i = 1,2,3$ leading to a total field of the form:

$$e = e^{\omega 1}(z,t) + e^{\omega 2}(z,t) + e^{\omega 3}(z,t)$$

If we substitute this into the wave equation we can separate the resulting equation into three equations, each containing only terms which oscillate at one of the three fundamental frequencies, and nonlinear polarization terms.
The nonlinar polarization terms will be of the form:

$$\mathrm{Re}\left[d^{(\omega_i \pm \omega_j)} E_1 E_2 e^{i[(\omega_i \pm \omega_j)t - (k_i \pm k_j)z]}\right]$$

and it's permutations. These terms oscillate at the sum and difference frequencies of the fundamental waves and will only be able to give rise to constructively interfering effects if we have a condition such as $\omega_3 = \omega_1 + \omega_2$.

We can now add the terms oscillating at the appropruiate frequencies to the three equations previously mentionned (i.e. maxwells equations for the linear case).

Making the slowly varying envelope approximation, $\left|k\frac{dE(z)}{\partial z}\right| >> \left|\frac{d^2E(z)}{dz^2}\right|$ and neglecting the dissipative terms we obtain three coupled mode equations:

$$\frac{dE_1}{dz} = -\frac{i\omega_1}{2}\sqrt{\frac{\mu}{\varepsilon_1}}dE_3E_2^* e^{-i(k_3 - k_1 - k_2)z}$$

$$\frac{dE_2^*}{dz} = \frac{i\omega_2}{2}\sqrt{\frac{\mu}{\varepsilon}}dE_1E_3^* e^{-i(k_1 - k_3 + k_2)z}$$

$$\frac{dE_3}{dz} = \frac{-i\omega_3}{2}\sqrt{\frac{\mu}{\varepsilon}}dE_1E_2 e^{-i(k_1 + k_2 - k_3)}$$

These are the basic equations describing second order nonlinear parametric interactions. They describe sum and difference frquency generation as well as parametric generation in which an incident high frequency wave generates two lower frequency waves, called signal and idler waves. This phenomena can be used to realize parametric oscillators which are capble of providing tunable coherent light over very large frequency bands. In coming years it is probable that parametric oscillators will be as ubiquitous as lasers are today. These sources also have fascinating implications for future "applications" such as quantum cryptography, quantum teleportation, quantum computing, etc.