Nonlinear effects

We've taken great advantage of linearity in developing our treatment of EM wave propagation in media. This is not always legitimate.

In fact, there are a number of nonlinear phenomena that are very general, quite important in optical communications, and possibly relevant in nanostructured photonic applications.

Here we'll very briefly look at:

- Origins of nonlinear optical effects
- Various kinds of nonlinear effects and where they come from, from a general 1d nonlinear oscillator picture
- Specific technologically relevant effects: solitons, optical Kerr effect, and the electro-optic effect.

General nonlinear model and definitions

In our original definition of $\varepsilon(\omega)$ and the Kramers-Kronig relations, we used the idea that the polarization ***P*** of a medium could only depend on the electric field at earlier times.

However, we only took the *linear* approximation of the most general kind of relationship. That is, we assumed:

$$\mathbf{P}_\alpha(\mathbf{r},t) = \varepsilon_0 \sum_\beta \int_{\mathbf{r}_1}\int_{t_1} dt_1 d\mathbf{r}_1 \chi_{\alpha\beta}(\mathbf{r}-\mathbf{r}_1, t-t_1)\mathbf{E}_\beta(\mathbf{r}_1,t_1)$$

Then our definition of $\varepsilon(\omega)$ is simply related to the Fourier transform of the linear susceptibility tensor $\chi_{\alpha\beta}$.

We could continue to include higher order terms:

$$\mathbf{P}_\alpha^{NL}(\mathbf{r},t) = \varepsilon_0 \sum_{\beta\gamma} \int_{\mathbf{r}_1}\int_{\mathbf{r}_2}\int_{t_1}\int_{t_2} d\mathbf{r}_1 d\mathbf{r}_2 dt_1 dt_2 \chi^{(2)}_{\alpha\beta\gamma}(\mathbf{r}-\mathbf{r}_1, t-t_1; \mathbf{r}-\mathbf{r}_2, t-t_2)\mathbf{E}_\beta(\mathbf{r}_1,t_1)\mathbf{E}_\gamma(\mathbf{r}_2,t_2)$$
$$+ \varepsilon_0 \sum_{\beta\gamma\delta} \int_{\mathbf{r}_1}\int_{\mathbf{r}_2}\int_{\mathbf{r}_3}\int_{t_1}\int_{t_2}\int_{t_3} d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 dt_1 dt_2 dt_3 \chi^{(3)}_{\alpha\beta\gamma\delta}(\mathbf{r}-\mathbf{r}_1, t-t_1; \mathbf{r}-\mathbf{r}_2, t-t_2; \mathbf{r}-\mathbf{r}_3, t-t_3)\mathbf{E}_\beta(\mathbf{r}_1,t_1)\mathbf{E}_\gamma(\mathbf{r}_2,t_2)\mathbf{E}_\delta(\mathbf{r}_3,t_3)$$

General nonlinear model and definitions

Appropriately defining with Fourier transforms:

$$\chi^{(2)}_{\alpha\beta\gamma}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2) = \int_{\mathbf{r}_1}\int_{\mathbf{r}_2}\int_{t_1}\int_{t_2} d\mathbf{r}_1 d\mathbf{r}_2 dt_1 dt_2 \exp[-i\mathbf{k}_1\cdot\mathbf{r}_1]\exp[+i\omega_1 t_1] \times \exp[-i\mathbf{k}_2\cdot\mathbf{r}_2]\exp[+i\omega_2 t_2]\chi^{(2)}_{\alpha\beta\gamma}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2)$$

$$\chi^{(3)}_{\alpha\beta\gamma}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2;\mathbf{k}_3,\omega_3) = \int_{\mathbf{r}_1}\int_{\mathbf{r}_2}\int_{\mathbf{r}_3}\int_{t_1}\int_{t_2}\int_{t_3} d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 dt_1 dt_2 dt_3 \exp[-i\mathbf{k}_1\cdot\mathbf{r}_1]\exp[+i\omega_1 t_1] \times \exp[-i\mathbf{k}_2\cdot\mathbf{r}_2]\exp[+i\omega_2 t_2]\exp[-i\mathbf{k}_3\cdot\mathbf{r}_3]\exp[+i\omega_3 t_3] \times \chi^{(3)}_{\alpha\beta\gamma\delta}(\mathbf{r}_1,t_1;\mathbf{r}_2,t_2;\mathbf{r}_3,t_3)$$

Clearly this gets even uglier if we go to even higher orders.

The important thing to bear in mind is which susceptibilities mix which frequencies.

General nonlinear 1d oscillator

We can at least see where various nonlinear effects come from by considering a nonlinear harmonic oscillator (and thinking of this as the nonlinear polarizing part of the medium, responding to the external EM field):

$$\ddot{x} + \Gamma\dot{x} + \omega_0^2 x + ax^2 + bx^3 + \cdots = \frac{q}{m}E(t)$$

The plan of attack is not too different from what you've seen in perturbation theory. Assume a solution of the form:

$$x(t) = x^{(1)}(t) + x^{(2)}(t) + x^{(3)}(t) + ...$$

where $x^{(n)}(t)$ is proportional to the nth power of the electric field. We can plug this in and group like powers of field to find:

$$\ddot{x}^{(1)}(t) + \Gamma\dot{x}^{(1)}(t) + \omega_0^2 x^{(1)}(t) = \frac{q}{m}E(t)$$

$$\ddot{x}^{(2)}(t) + \Gamma\dot{x}^{(2)}(t) + \omega_0^2 x^{(2)}(t) + a(x^{(1)}(t))^2 = 0$$

$$\ddot{x}^{(3)}(t) + \Gamma\dot{x}^{(3)}(t) + \omega_0^2 x^{(3)}(t) + 2ax^{(1)}(t)x^{(2)}(t) + b(x^{(1)}(t))^3 = 0$$

$$\vdots$$

General nonlinear 1d oscillator

We can rewrite this in the frequency domain. We'll do the first and second order pieces, and then just state the results for higher orders. Assuming the usual sort of harmonic solution for $x^{(1)}$,

$$x^{(1)}(\omega) = \frac{q}{m} E(\omega) \cdot \underbrace{\left[\omega_0^2 - \omega^2 - i\omega\Gamma\right]^{-1}}_{F(\omega_0, \omega; \Gamma)}$$

The second order term looks like:

$$\ddot{x}^{(2)}(t) + \Gamma \dot{x}^{(2)}(t) + \omega_0^2 x^{(2)}(t) = -a(x^{(1)}(t))^2$$
$$x^{(2)}(\omega) = -aF(\omega_0, \omega; \Gamma) \int d\omega' x^{(1)}(\omega') x^{(1)}(\omega - \omega')$$

Plugging in,

$$x^{(2)}(\omega) = -a\left(\frac{q}{m}\right)^2 F(\omega_0, \omega; \Gamma) \int d\omega' F(\omega_0, \omega'; \Gamma) F(\omega_0, (\omega - \omega'); \Gamma) E(\omega') E(\omega - \omega')$$

General nonlinear 1d oscillator

We can continue this for higher orders. The interesting thing that happens is when we consider driving terms composed of two different frequencies:

$$E(t) = E_1 \exp(-i\omega_1 t) + E_2 \exp(-i\omega_2 t)$$

Plugging this in, because of the nonlinearity mixing results, producing polarization response at frequencies other than just the driving frequencies ω_1 and ω_2.

Resulting types of phenomena

Among the effects that result:

- Second harmonic generation
- Sum frequency generation
- Difference frequency generation
- “Optical rectification”
- Third harmonic generation
- Intensity dependent propagation
- Raman generation
- Electro-optic effect

We’ll very briefly look at a couple of these to see where theycome from, but look in more detail at those emphasized above.

Second harmonic generation

Plugging in our two frequency driving term into our order-by-order hierarchical solution to the nonlinear oscillator, we end up with polarization contributions at a number of frequencies. One example is second harmonic generation. Because of the cubic nonlinearity (basically $\chi^{(2)}$), we get contributions:

$$x^{(2)}(2\omega_1) = -a\left(\frac{q}{2m}\right)^2 F(\omega_0, 2\omega_1; \Gamma) F^2(\omega_0, \omega_1; \Gamma) \times E_1^2(\omega_1)$$

$$x^{(2)}(2\omega_2) = -a\left(\frac{q}{2m}\right)^2 F(\omega_0, 2\omega_2; \Gamma) F^2(\omega_0, \omega_2; \Gamma) \times E_2^2(\omega_2)$$

Sum and difference generation

Similarly, the third order nonlinearity (second order susceptibility) leads to material polarization at the sum and difference frequencies, as well as a dc component!

That is, there are terms like:

$$x^{(2)}(\omega_1+\omega_2) = -a\left(\frac{q}{2m}\right)^2 F(\omega_0,\omega_1+\omega_2;\Gamma)F(\omega_0,\omega_1;\Gamma)F(\omega_0,\omega_2;\Gamma)\times E_1(\omega_1)E_2(\omega_2)$$

$$x^{(2)}(\omega_1-\omega_2) = -a\left(\frac{q}{2m}\right)^2 F(\omega_0,\omega_1-\omega_2;\Gamma)F(\omega_0,\omega_1;\Gamma)F^*(\omega_0,\omega_2;\Gamma)\times E_1(\omega_1)E_2^*(\omega_2)$$

$$x^{(2)}(0) = -a\left(\frac{q}{2m}\right)^2 F(\omega_0,0;\Gamma)\left|F(\omega_0,\omega_1;\Gamma)\right|^2\times\left|E_1(\omega_1)\right|^2$$

$$x^{(2)}(0) = -a\left(\frac{q}{2m}\right)^2 F(\omega_0,0;\Gamma)\left|F(\omega_0,\omega_2;\Gamma)\right|^2\times\left|E_2(\omega_2)\right|^2$$

Effects of 3rd order NL susceptibility

The quartic term in the potential (3rd order NL susceptibility piece) leads to three more interesting effects.

- Third harmonic generation - there are terms in $x^{(3)}(\omega)$ that depend on driving frequencies like $3\omega_1$ and $3\omega_2$.
- Intensity-dependent propagation - we explore this more on the next slide.
- Raman generation - inelastic process that we won't discuss in detail.

Intensity dependent propagation

Simplest to see just by plugging into the 1d model directly:

$$\ddot{x}+\Gamma\dot{x}+\omega_0^2 x+bx^3=\frac{q}{2m}E_0[e^{i\omega t}+e^{-i\omega t}]$$

Suppose the nonlinearity is a small effect. Then the solution $x(t)$ should look a lot like the simple linear case:

$$x(t)\approx\frac{1}{2}[x_0 e^{i\omega t}+x_0 * e^{-i\omega t}]$$

Plugging this in for x and looking at the 3rd order term we get:

$$bx^3(t)=\frac{3}{8}b|x_0|^2[x_0 e^{i\omega t}+x_0^* e^{-i\omega t}]+\frac{1}{8}b[x_0^3 e^{i3\omega t}+x_0^{*3} e^{-i3\omega t}]$$

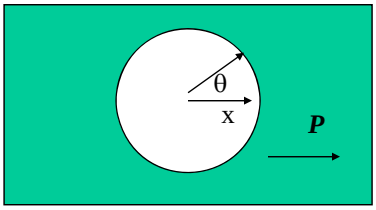
For small deviations from linearity, we find

$$|x_0|^2\approx\frac{q^2}{\left|m(\omega_0^2-\omega^2-i\Gamma\omega)\right|^2}|E_0|^2$$

The result is an effective index that depends on intensity! $n\approx n_L+n_2 I$

Claussius-Mossotti relation

To better see the link between this nonlinear charge displacement and index of refraction, we need to recall the Claussius-Mossotti relation.



If we define an atomic polarizability α, and we consider the relationship between local electric field and polarization,

$$E_x=\frac{-1}{4\pi\varepsilon_0 r^2}\int_0^{\pi}(-P\cos\theta)\cdot\cos\theta\cdot[2\pi r^2\sin\theta d\theta]=\frac{P}{3\varepsilon_0}$$

$$\mathbf{E}_{loc}=\mathbf{E}+\frac{\mathbf{P}}{3\varepsilon_0}\qquad \mathbf{P}=\sum_j N_j\alpha_j\mathbf{E}_{loc}=\varepsilon_0(\frac{\varepsilon}{\varepsilon_0}-1)\mathbf{E}\qquad \mathbf{P}=\left[\frac{\sum_j N_j\alpha_j}{1-\frac{1}{3\varepsilon_0}\sum_j N_j\alpha_j}\right]\mathbf{E}$$

Plugging into our definition of the dielectric constant, we find

$$\frac{\varepsilon}{\varepsilon_0}=n^2=\left[1+\frac{\frac{1}{\varepsilon_0}\sum_j N_j\alpha_j}{1-\frac{1}{3\varepsilon_0}\sum_j N_j\alpha_j}\right]\qquad \frac{n^2-1}{n^2+2}=\frac{1}{3\varepsilon_0}\sum_j N_j\alpha_j$$

Claussius-Mossotti relation

So, using our third order nonlinear relation for the displacement, we can find

$$\alpha(|E_0|^2) = \frac{q^2}{m\left[\omega_0^2 + \frac{3}{4}b|\alpha_L E_0^2| - \omega^2 - i\Gamma\omega\right]}$$

Typical numbers for some materials for n_2:

Glass fiber: 10^{-16} cm^2/W over most frequencies of interest
GaAs: 10^{-7} cm^2/W at 820 nm
InSb: 10^{-3} cm^2/W at 5 microns.

Downside of these later materials: significant losses.

Self-focusing

Start with your favorite wave equation. For example, consider the vector potential: $\nabla^2\mathbf{A} + \omega^2\mu_0\varepsilon_0 n(z)^2\mathbf{A} = 0$

Assume that ***A*** is in the *y* direction, the wave propagates in the *z* direction, and that it has some shape: $\mathbf{A} \sim \hat{\mathbf{y}}u(x,y,z)\exp(-ik_0 z)$

Normalize so that $|u|^2$ is the intensity, and define k_0 as the "unperturbed" wavevector magnitude, $k_0 = \omega\sqrt{\mu_0\varepsilon_0 n_0}$

Separating out longitudinal and transverse parts of the laplacian, and applying the chain rule,

$$\nabla_T^2 u - 2ik_0\frac{\partial u}{\partial z} = -\omega^2\mu_0\varepsilon_0(n^2 - n_0^2)u$$

In the 2d case,

$$\frac{\partial^2}{\partial x^2}u - 2ik_0\frac{\partial u}{\partial z} = -\omega^2\mu_0\varepsilon_0(n^2 - n_0^2)u$$

Self-focusing

Assuming our small third order nonlinearity that gives us

$$n \approx n_L + n_2 I = n_0 + n_2 |u|^2$$

we expand to first order in $n_2|u|^2$ and get

$$\frac{\partial^2}{\partial x^2} u - 2ik_0 \frac{\partial u}{\partial z} + 2k_0^2 \frac{n_2}{n_0} |u|^2 u = 0$$

Changing variables,

$$q \equiv \frac{z}{2k_0} \qquad \kappa \equiv 2k_0^2 \frac{n_2}{n_0}$$

we end up with an equation that may look quite familiar:

$$-i \frac{\partial}{\partial q} u = \left(-\frac{\partial^2}{\partial x^2} - \kappa |u|^2 \right) u$$

This is the nonlinear Schroedinger equation, with q playing the role of time, and $-\kappa|u|^2$ playing the role of the (nonlinear) potential.

Self-focusing

$$-i \frac{\partial}{\partial q} u = \left(-\frac{\partial^2}{\partial x^2} - \kappa |u|^2 \right) u$$

For positive κ (that is, $n_2 > 0$), this has a general solution:

$$u = \sqrt{\frac{2}{\kappa}} \eta \frac{\exp\left[i(\xi^2 - \eta^2) q + i\xi x - i\phi \right]}{\cosh\left[\eta(x - x_0) + 2\eta\xi q \right]}$$

In this solution, ξ, η, ϕ, and x_0 are arbitrary parameters. To see what this solution means, start by assuming ξ and $\phi = 0$.

$$u \exp(-ik_0 z) = \sqrt{\frac{2}{\kappa}} \eta \frac{\exp\left[-i\eta^2 q \right]}{\cosh\left[\eta(x - x_0) \right]} e^{-ik_0 z}$$

This is a beam with an x-dependent profile with width ~ $1 / \eta$.

The effective wavevector is modified from the linear case to

$$k = k_0 \left[1 + \left(\frac{\eta^2}{2k_0^2} \right) \right]$$

Self-focusing

$$u = \sqrt{\frac{2}{\kappa}}\eta \frac{\exp\left[i(\xi^2 - \eta^2)q + i\xi x\right]}{\cosh\left[\eta(x - x_0) + 2\eta\xi q\right]}$$

The solution with nonzero ξ describes a pulse inclined to the *z* axis.

One other interesting feature is that the integral $\sqrt{k/2}\int_{-\infty}^{\infty} |u|\,dx$ always equals π, independent of the beam parameters.

Why is this situation called self-focusing? The higher the intensity (the larger |u| is to begin with), the *narrower* the resulting steady state beam profile.

Intuitively, because of the nonlinearity the index goes up where the beam intensity is the greatest, leading to waveguiding-like behavior.

$n \sim n_0 + n_2|u|^2$
$n \sim n_0$

Self-focusing

That integral condition means typical self-focused beam always contains about the same amount of power, independent of beam parameters.

What happens if one starts off with too much power? No stable propagating solution: “Catastrophic” focusing to a point - results in damage in high powered laser systems.

I’ll try to guide you through a self-focusing problem on the problem set.

Solitons

Another effect of this kind of nonlinearity is the existence of solutions that are called *solitons*.

Consider a propagating mode confined to an optical fiber, with an effective wavevector $k(\omega)$ propagating in the z direction. With an appropriate change of variables: $\tau \equiv t - \dfrac{z}{(\partial\omega/\partial k)}$

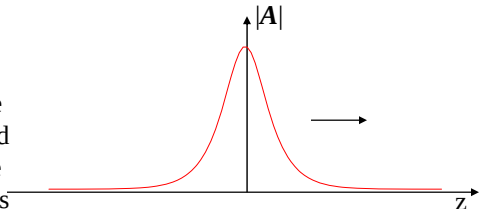
The vector potential must obey:

$$i\frac{\partial}{\partial z}\mathbf{A} + \frac{1}{2}\frac{\partial^2 k}{\partial\omega^2}\frac{\partial^2 \mathbf{A}}{\partial\tau^2} - \kappa|\mathbf{A}|^2\mathbf{A} = 0 \qquad \kappa \equiv \frac{\omega_0^2\mu_0\varepsilon_0}{k(\omega_0)}\frac{\int n_0 n_2 |u|^4 da}{\int |u|^2 da}$$

The solutions to this wave equation are solitons, provided there's anomalous dispersion: $\dfrac{\partial^2 k}{\partial\omega^2} < 0$

Solitons

Imagine sending in an optical pulse with some carrier frequency ω_0, and assume for a moment that the pulse bandwidth is small compared to this carrier.



Solution is a soliton, a pulse with a hyperbolic secant shape. The envelope propagates at the group velocity at the carrier frequency.

Basically, the nonlinearity, which tries to compress the pulse (self-focusing tendency), compensates for the dispersion, which tries to spread the pulse.

Result is dispersion-free pulse propagation.

Solitons in communications

This dispersion-free propagation was recognized a long time ago as something potentially useful for communications.

However, things get complicated when one tries to implement this at high bandwidths, and solitons have remained largely in the laboratory.

This may be changing, though, as nanoscale control over materials properties + photonic band gap fibers, etc. make possible more tuning of parameters.

Sense of sizes of things: in high bandwidth lab tests, typical pulse durations of 1.55 micron light are ~ 10 ps. That's about 2000 periods of oscillation.

Optical Kerr effect

Another unintuitive result of that 3rd order nonlinear susceptibility: if the intensity results from two counter-propagating waves, each wave sees a *different* effective index.

This comes from: $\mathbf{P} \approx \chi_l \mathbf{E} + \chi_{nl} |\mathbf{E}|^2 \mathbf{E} = \chi_l \mathbf{E} + \chi_{nl} \mathbf{E}\mathbf{E}^* \mathbf{E}$

$$\mathbf{E} = \left[\mathbf{E}_+ \exp(ik_+ z) + \mathbf{E}_- \exp(-ik_- z)\right] e^{-i\omega t}$$

$$k_+ = k_0 \left[n_0^2 + \chi_{nl} \frac{|\mathbf{E}_+|^2 + 2|\mathbf{E}_-|^2}{\varepsilon_0} \right]^{1/2}$$

$$k_- = k_0 \left[n_0^2 + \chi_{nl} \frac{|\mathbf{E}_-|^2 + 2|\mathbf{E}_+|^2}{\varepsilon_0} \right]^{1/2}$$

$$\longrightarrow \quad n_+ = n_0 + n_2 |\mathbf{E}_+|^2 + 2n_2 |\mathbf{E}_-|^2$$

$$n_- = n_0 + n_2 |\mathbf{E}_-|^2 + 2n_2 |\mathbf{E}_+|^2$$

This actually can be a dominant source of error in optical gyroscopes.

Electro-optic effect

$$\ddot{x}+\Gamma\dot{x}+\omega_0^2 x+ax^2+bx^3+\cdots=\frac{q}{m}E(t)$$

So far we've been worrying about the *b* term here.

Turns out that the *a* term can only realistically exist in a solid with a broken inversion symmetry.

The *a* term actually leads to some very useful consequences. Neglect *b* and consider the effect of a *dc* electric field in addition to our usual ac drive:

$$\ddot{x}+\Gamma\dot{x}+\omega_0^2 x+ax^2=\frac{q}{m}\left[\frac{E_0}{2}(e^{i\omega t}+e^{-i\omega t})+E_{dc}\right]$$

We're interested in the effect of the dc field on the ac response, so assume a solution (neglecting h.o.t.) of the form:

$$x(t)=x_0+\frac{1}{2}(x_1 e^{i\omega t}+x_1^* e^{-i\omega t})$$

Electro-optic effect

Plugging in, we get for the dc terms:

$$\frac{1}{2}\omega_0^2 x_0+a\left[x_0^2+\frac{|x_1|^2}{2}\right]=\frac{q}{m}E_{dc}$$

and for the ac terms,

$$x_1\left[\omega_0^2-\omega^2-i\Gamma\omega+2ax_0\right]=\frac{q}{m}E_0$$

The end result is an effective index that depends upon the dc field.

Think about it this way: oscillations of charges around equilibrium positions give the index of refraction. Because of the nonlinearity, the frequency response of the oscillations can be shifted by applying a dc field (and changing the no-drive equilibrium position of the charges).

Electro-optic effect

In realistic 3d materials, one can define principle axes of symmetry, and choose coordinates in which to write down the full tensorial dielectric response:

$$\mathbf{D} = \tilde{\varepsilon}\mathbf{E} \quad \longrightarrow \quad \mathbf{E} = \tilde{\eta}\mathbf{D} = \frac{1}{\varepsilon_0}\begin{bmatrix} 1/n_x^2 & 0 & 0 \\ 0 & 1/n_y^2 & 0 \\ 0 & 0 & 1/n_z^2 \end{bmatrix}\mathbf{D}$$

A dc field modifies this tensor:

$$\mathbf{E} = \left(\frac{1}{\varepsilon_0}\begin{bmatrix} 1/n_x^2 & 0 & 0 \\ 0 & 1/n_y^2 & 0 \\ 0 & 0 & 1/n_z^2 \end{bmatrix} + \begin{bmatrix} \delta\eta_1 & \delta\eta_6 & \delta\eta_5 \\ \delta\eta_6 & \delta\eta_2 & \delta\eta_4 \\ \delta\eta_5 & \delta\eta_4 & \delta\eta_3 \end{bmatrix} \right)\mathbf{D}$$

where

$$\delta\tilde{\eta} = \tilde{\mathbf{r}}\mathbf{E}_{dc} = \begin{bmatrix} r_{1x} & r_{1y} & r_{1z} \\ \vdots & \vdots & \vdots \\ r_{6x} & r_{6y} & r_{6z} \end{bmatrix}\mathbf{E}_{dc}$$

Electro-optic effect

These *r*s are called electro-optic coefficients.

To summarize: an electro-optic coefficient relates the change in $(1/\varepsilon_0 n^2)$ in some propagation direction to an applied dc electric field in a different direction (!).

How big are some of these numbers?

For GaAs, $r_{4x} = r_{5y} = r_{6z} = 5.9 \times 10^{-12}$ m/V.

Assume propagation in the *z* direction, and an applied electric field in the *x* direction. The result is a change in the index for modes with the electric field along the *y* direction, and the size of the effect is about:

$$\delta n \approx -\frac{1}{2} n^3 r E_{dc-x}$$

For light at 900 nm, $n = 3.6$, and assuming 10^7 V/m, the index changes by around 0.0013.

Electro-optic effect

This is about as large as these effects get. The problem is usually that electro-optic coefficients are small, and that breakdown dc fields for materials with large electro-optic coefficients are poor.

The industry favorite for much of these effects: $LiNbO_3$, which has coefficients 4-5 times larger than GaAs.

We'll see later why having large EO effects is desirable.

Also note: systems with big EO coefficients have lots of charge arranged in a nonsymmetric way in the unit cell. That tends to lead to other effects that can be important (*e.g.* piezoelectricity, pyroelectricity, etc.).

Again, nanoscale structuring and tuning of material properties to enhance desired ones without introducing other problems is a very active area of research.

Acousto-optic effect

One other effect I want to mention is the acousto-optic (AO) effect. It's not a nonlinearity per se, but now's a good time to see where it comes from.

Recall the derivation of the Clausius-Mossotti relation:

$$\varepsilon = n^2 = \left[1 + \frac{\frac{1}{\varepsilon_0}\sum_j N_j \alpha_j}{1 - \frac{1}{3\varepsilon_0}\sum_j N_j \alpha_j} \right]$$

Those *N*s are numbers of polarizable objects per unit volume.

Longitudinal sound waves correspond to local density modulations of the medium.

Therefore, sound waves can periodically modulate the index of refraction, on a length scale corresponding to the acoustic wavelength of the medium!

We'll do a problem on this….

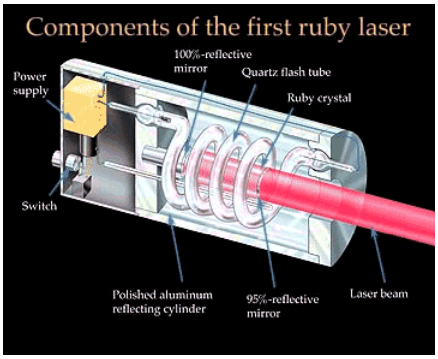
Summary and next time

- Anharmonic behavior of the polarizability leads to nonlinear behavior of optical media.
- The result can be all kinds of frequency mixing effects, as well as couplings between intensity and propagation direction.
- Those latter effects, involving the 3rd order NL susceptibility, give rise to things like self-focusing and solitons.
- Microscopic material properties like lack of inversion symmetry can give rise to other profound effects.

Next time: lasers

Lasers

- Brief look at essential physics of matter and radiation
- Laser idea and rate equation approach
- Semiclassical theory description and results
- Some common lasing systems



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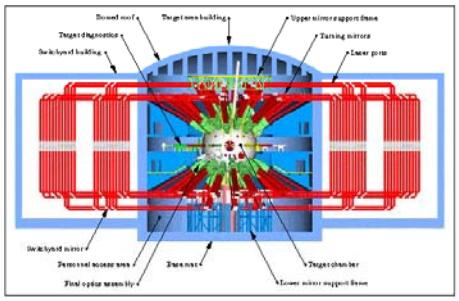


NIF, LLNL

JPL

Time-dependent perturbation theory

When we consider the interactions between radiation and matter, we're interested in the response of a system to a perturbing Hamiltonian that varies harmonically with time.

Appropriate description: time-dependent perturbation theory.

Assume unperturbed system (the matter) can be represented by a complete set of eigenstates: $H_0\left|\psi_j^0\right\rangle = E_j^0\left|\psi_j^0\right\rangle$

Also assume for now that these states are not degenerate. We should always be able to expand the time-dependent solution in terms of these eigenstates, with appropriate time-dependent coefficients: $\left|\Psi(t)\right\rangle = \sum_j c_j(t)e^{-iE_j^0 t/\hbar}\left|\psi_j^0\right\rangle$

Plugging into time-dependent Schroedinger equation with perturbation $H'(t)$, and taking inner product with unperturbed state,

$$i\hbar\dot{c}_k(t) = \sum_j c_j(t)\left\langle\psi_k^0\left|H'(t)\right|\psi_j^0\right\rangle e^{i(E_k^0-E_j^0)t/\hbar}$$

Time-dependent perturbation theory

Defining $\omega_{kj} \equiv \frac{E_k^0 - E_j^0}{\hbar}$ $\qquad H'_{kj} \equiv \left\langle \psi_k^0 \middle| H' \middle| \psi_j^0 \right\rangle$

Let's assume the system started out in unperturbed state *a*. We can then solve the differential equation for a generic coefficient $c_j(t)$ to ask what is the probability of finding the system in state *j* at some later time.

$$c_j(t) = \frac{1}{i\hbar}\int_0^t H'_{ja}(\tau) e^{i\omega_{ja}\tau} d\tau$$

$$P_{ja}(t) = \left|c_j(t)\right|^2 = \frac{1}{\hbar^2}\left|\int_0^t H'_{ja}(\tau) e^{i\omega_{ja}\tau} d\tau\right|^2$$

For a harmonic perturbation, $H'(t) = \frac{V_0}{2}\left(e^{i\omega t} + e^{-i\omega t}\right)$

we find:

$$c_j(t) = -i\frac{V_{0,ja}}{\hbar}\left[\frac{e^{i(\omega_{ja}+\omega)t} - 1}{i(\omega_{ja}+\omega)} + \frac{e^{i(\omega_{ja}-\omega)t} - 1}{i(\omega_{ja}-\omega)}\right]$$

Time-dependent perturbation theory and stimulated emission

Define $F(t,\Omega) \equiv \frac{2\sin^2 \Omega t/2}{\Omega^2}$

We find that the transition probability is

$$P_{ja}(t) = F(t,\omega_{ja}+\omega)\left|V_{0,ja}\right|^2 + F(t,\omega_{ja}-\omega)\left|V_{0,ja}\right|^2 + \text{ cross terms}$$

At long times *F* acts like $F(t,\Omega) \to \pi t\,\delta(\Omega)$

So, at long times only transitions that absorb or emit an amount of energy $\hbar\omega$ are permitted.

The result: first term represents *stimulated emission*.

Second term: *absorption*.

- Actual transition rates will depend on matrix element of perturbation between initial and final states.
- Density of final states available will also contribute to rate (as will statistics of particles).

Interaction with radiation

Simplest type of perturbation when we're discussing matter and radiation is the *electric dipole* interaction. For this case, if our unperturbed states are electronic eigenstates, the perturbation term really looks like: $H'(t) = \mathbf{E}_0 \cdot (-e\mathbf{r}) \cos \omega t$

If θ is the angle between the **E** field polarization and the dipole moment, we can write the absorption rate as:

$$W_{abs} = \frac{\pi I(\omega)}{\hbar^2 c \varepsilon_0} \cos^2 \theta \left| \mathbf{p}_{ja} \right|^2$$ where I is intensity of radiation,

$$\mathbf{p}_{ja} \equiv \left\langle \psi_j \right| -e\mathbf{r} \left| \psi_a \right\rangle \qquad \mathbf{p}_{ja} = \mathbf{p}^*_{aj} \rightarrow \left| \mathbf{p}_{ja} \right|^2 = \left| \mathbf{p}_{aj} \right|^2$$

Note that because of this last equation, the stimulated emission rate is identical to the absorption rate, under the influence of the same radiation. This is called *detailed balance*.

Spontaneous emission and Einstein A and B coefficients

Of course, there is some rate for excited atoms (for example) to emit radiation, independent of any external field. This is spontaneous emission.

In some cavity, if there are N_a atoms in the ground state and N_b atoms in an excited state, and an energy density $\rho(\omega_{ab})$ of radiation at the appropriate frequency to cause transitions, the rate of absorption transitions is:

$$\dot{N}_{ba} = B_{ba} N_a \rho(\omega_{ab})$$

From the previous slide, we can identify where we've averaged over θ. $B_{ba} = \frac{\pi}{3\hbar^2 \varepsilon_0} \left| \mathbf{p}_{ba} \right|^2$

Emission rate is given by: $\dot{N}_{ab} = A_{ab} N_b + B_{ab} N_b \rho(\omega_{ab})$

Here A_{ab} is the Einstein coefficient for spontaneous emission.

In equilibrium the two transition rates have to be equal.

Einstein A and B coefficients

We also know what the ratio has to be between excited and ground state populations in thermal equilibrium:

$$\frac{N_a}{N_b} = \exp(\hbar\omega_{ab} / k_B T) = \frac{A + B\rho(\omega_{ab})}{B\rho(\omega_{ab})}$$

We can solve this for the radiation energy density.

If we compare the resulting formula with the general Planck spectrum for a 3d cavity in thermal equilibrium, we can find A in terms of B, and quickly solve for the spontaneous emission rate:

$$W_{spont} = \frac{\omega_{ab}^3}{3\pi c^3 \hbar \varepsilon_0} |\mathbf{p}_{ab}|^2$$

Notes:

- In equilibrium, we can never get population inversion (would require negative temperature).
- If our system is in a cavity with a restricted photon density of states, we can actually modify the spontaneous emission rate!

Cross-sections

Many treatments of light-matter interactions speak in terms of cross-sections.

Just to refresh your memory, $\sigma \equiv \frac{W\hbar\omega}{I}$

That is, the total cross-section for some process that emits energy in chunks of $\hbar\omega$ is given by the per particle rate of energy emission ($W\,\hbar\omega$) divided by the incident intensity (energy per unit area per unit time).

The LASER idea

"Light amplification by the stimulated emission of radiation".

Consider radiation at frequency ω coming into a material filled with two-level systems with energy splittings resonant with that frequency.

The radiation energy density change from absorption: $\dot{\rho} = -N_1 \hbar \omega W_{abs}$

From stimulated emission: $\dot{\rho} = +N_2 \hbar \omega W_{stim}$

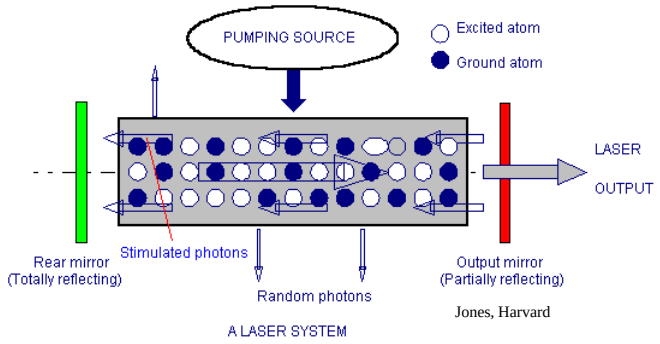
The net change is proportional to N_2-N_1, the difference between excited and ground state populations.

The idea of the laser is to get net gain for the radiation energy density by somehow having *population inversion*.

Note that *coherence* actually results from the boson nature of photons! Emission tends to go into modes already populated….

Caveat: one can actually get lasing *without* inversion, if one cleverly stacks the deck to suppress absorption (needs a third level).

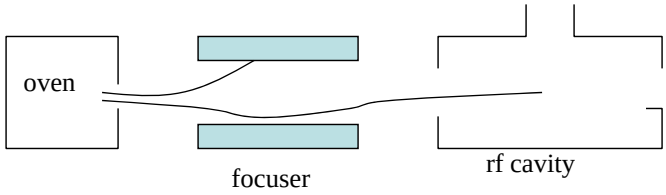
A generic laser:



- A pump source produces population inversion.
- The optical cavity makes those photons which occupy it's modes stick around much longer than other photons.
- Result is they have many more opportunities to cause spontaneous emission.
- Light is coupled out through leaky part of cavity.

Can get 2-level lasing….

Basic idea is the ammonia maser (emits in the microwave):



Take usual thermal distribution, but somehow (nonthermodynamically) lop off the ground state population (say, by magnetically selecting them out).

Result is a beam that's mostly excited molecules. Note that this does not work if you want to work with a fixed population….

This is basically the way semiconductor lasers work, though….

Why do you need at least 3 levels?

Steady-state population inversion by purely optical pumping can't be achieved in a purely two-level system.

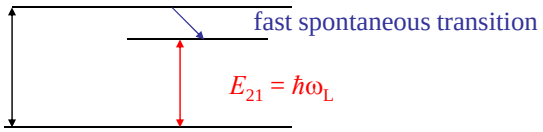
Can treat this mathematically (see http://www.stanford.edu/class/ee231/LectureNotes/ for lecture 11, for a nice treatment).

The physical picture: because absorption and stimulated emission come from the same physics, you can never get more energy out steady-state for the beam than you put in.

3- and 4-level systems

Basic idea for a 3-level system:

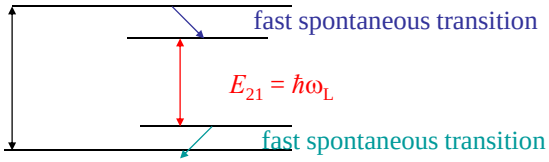
$E_{31} = \hbar\omega_p$



Each of these transitions has its own stimulated (and absorption) rates (Einstein *B* coefficients) and its own spontaneous rates (*A* coeff.).

Can pump at one frequency continuously to maintain population inversion between lower two levels.

Similarly, for 4-level case,



Master equations

One can write down master equations that express the constraints connecting the rates of all the various processes as these systems interact with radiation.

We won't go through this in excruciating detail (read Jones lecture notes or Stanford lecture notes if you're interested).

Look at 3-level case. If spontaneous transition rates W_{spont} are rewritten as γ, and ignoring stim. emission of the lasing level,

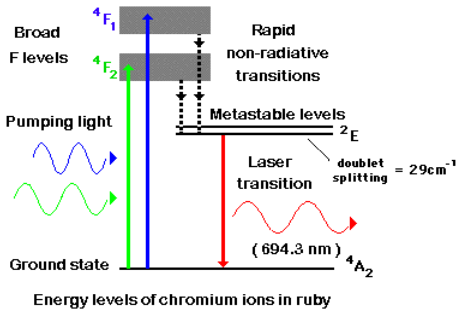
Top level: $\frac{dN_3}{dt} = W_p(N_1 - N_3) - N_3\gamma_3 \qquad \gamma_3 >> W_p \rightarrow N_3 \approx \frac{W_p}{\gamma_3} N_1$

lasing level: $\frac{dN_2}{dt} \approx N_3\gamma_3 - N_2\gamma_2 \longrightarrow N_2 \approx \frac{W_p}{\gamma_2} N_1$

So, to get steady state inversion, need $W_p > \gamma_2$.

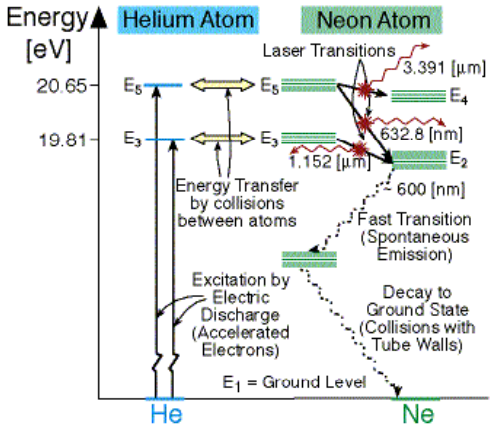
In practice, it's worse than this because stim. emission lowers inversion.

Some common laser systems:



Chromium ion (Cr^{+3}) impurities in crystalline Al_2O_3 - the basic ruby laser. Effectively a 3-level laser.

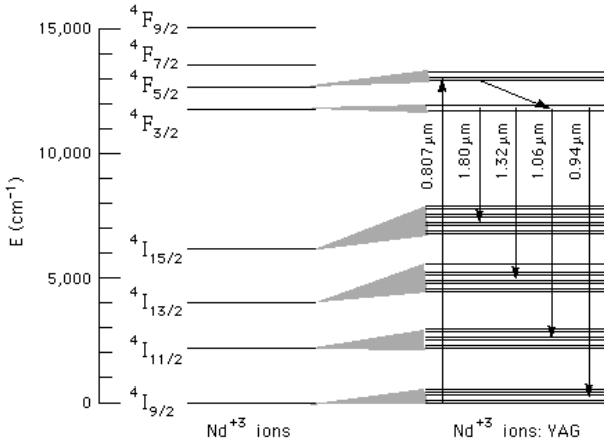
Helium-Neon



Electric spark pumps helium into long-lived states; collisional energy transfer excites the neon atoms into laser-active levels.

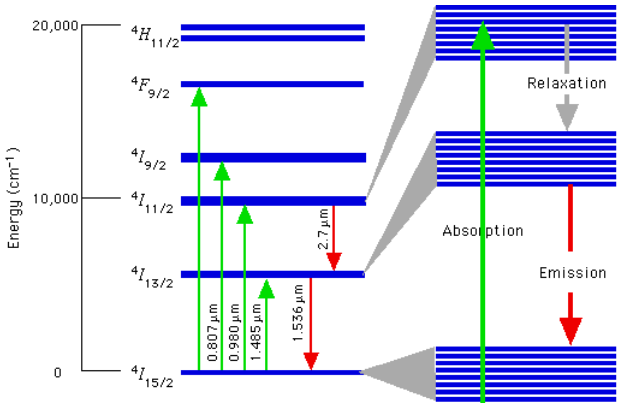
Effectively a 4-level laser.

Nd-YAG



Rare earth ions with transitions of convenient frequencies can be doped into optically boring (i.e. transparent) hosts. Example: neodymium ions in yttrium garnet ($Y_3Al_5O_{12}$).

Erbium in glass



Another technologically useful example: Er^{+3} ions in silicate glass. The basis for fiber amplifiers.

General point

As light at frequency of interest propagates in the gain medium, there is a competition between *absorption* losses and *gain*.

The point where pumping is effective enough that emission just balances absorption is called the threshold for transparency.

I strongly encourage you to at least skim the Jones lecture notes and the Stanford notes as well, to get a better feel for the requirements to actually get gain, and calculating thresholds and so forth.