Nonlinear Optics (WiSe 2017/18)

Lecture 11: November 23, 2017

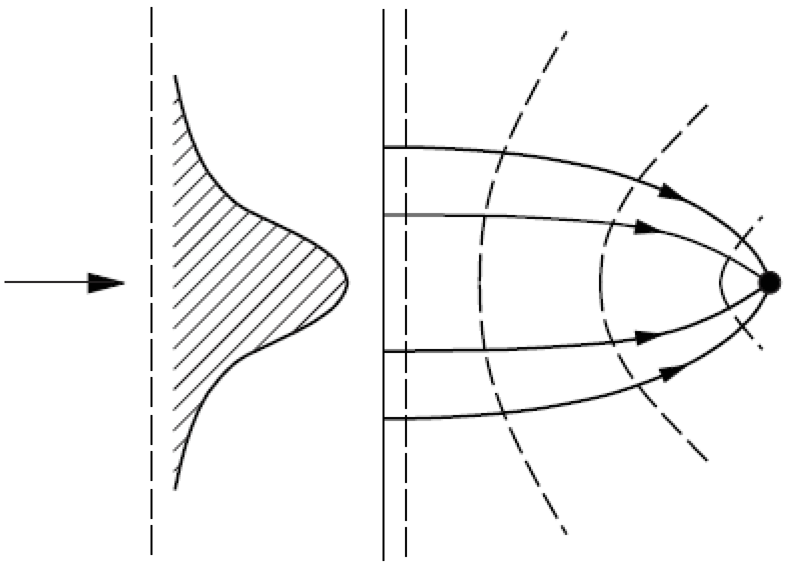
7 Third-order nonlinear effects (continued)

7.5 Self-focusing

7.6 Raman and Brillouin scattering

7.5 Self-focusing

transverse beam profile becomes instable



intensity-dependent refractive index

for $\Delta n_2 > 0$:

- phase velocity in center reduced
- phase fronts bend due to the induced lens ("**Kerr lens**")
- self-focusing of the beam

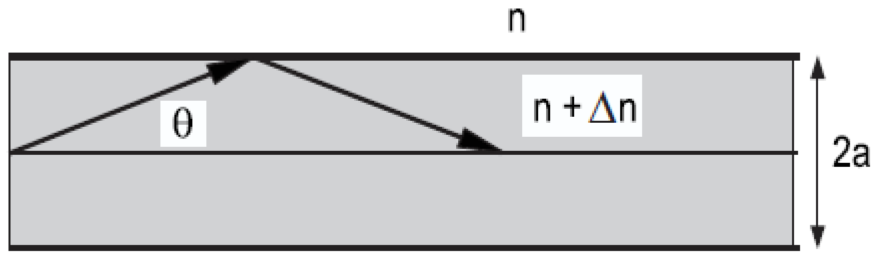
R. Y. Chiao, E. Garmire, and C. H. Townes, Phys. Rev. Lett. **13**, 479 (1964).

H. A. Haus, Appl. Phys. Lett. **8**, 128 (1966).

relevance:

- Kerr-lens mode-locked laser oscillators
- unwanted detrimental effect of "hot spots"

simple physical consideration in 2D:



Snell's law → total internal reflection for $\theta < \theta_c$, with $\cos\theta_c = \frac{n}{n+\Delta n}$

$$\cos\theta_c \cong 1 - \frac{\theta_c^2}{2} \cong 1 - \frac{\Delta n}{n} \Rightarrow \theta_c \cong \sqrt{2\Delta n/n}. \tag{7.46}$$

If a beam of diameter $2a$ propagates through the medium, it contains, because of diffraction, rays with an angle

$$\theta_B = \left(\frac{\pi/a}{k_0 n}\right) = \frac{\lambda/n}{2a}. \tag{7.47}$$

The refractive index difference, for which all these rays are totally reflected (i.e., trapped) then follows from $\theta_c = \theta_B$, thus

$$\Delta n_c = n_2^I I_c = \frac{\theta_c^2}{2} n. \tag{7.48}$$

From this we obtain, independent of the beam diameter, a critical power of the beam

$$P_c = \frac{\pi\lambda^2}{8nn_2}. \tag{7.49}$$

$$P_c = \frac{\pi\lambda^2}{8nn_2}. \qquad (7.49)$$

above this critical power, self-focusing exceeds diffraction.

Note the **quadratic scaling with wavelength**!

in 2D (1 longitudinal, 1 transversal dimension): **spatial solitons** occur.

in 3D (2 transversal dimensions):
catastrophic self-focusing occurs, that eventually is balanced by other nonlinear effects, e.g.,
- saturation of the intensity-dependent refractive index
- self-defocusing due to plasma formation by multi-photon ionization (**"filamentation"**)

A. Couairon and A. Mysyrowicz, Phys. Reports **441**, 47 (2007).

L. Bergé, S. Skupin, R. Nuter, J. Kasparian, and J.-P. Wolf, Rep. Prog. Phys. **70**, 1633 (2007).

Photograph of a self-guided filament induced in air by a high-power infrared (800 nm) laser pulse   [from http://www.teramobile.org]

Remote detection of biological aerosols. The tube in the center of the picture is an open cloud chamber generating the bioaerosol simulant. The laser beam is arriving from the left. [from http://www.teramobile.org]

High-voltage lightning: (left) without laser guiding, (right) with laser guiding.
[from http://www.teramobile.org]

In the paraxial approximation i.e., small-angle approximation

$$k_z = \sqrt{k^2 - \left(k_x^2 + k_y^2\right)} \approx k - \frac{1}{2k}\left(k_x^2 + k_y^2\right). \tag{7.50}$$

The dispersion relation within the paraxial approximation reads

$$\frac{\omega}{c} - k_z - \frac{1}{2k}\left(k_x^2 + k_y^2\right) = 0. \tag{7.51}$$

Taylor expansion around the carrier wave with carrier frequency and wave number in z-direction (ω_0, k_0), i.e., $\omega = \omega_0 + \Delta\omega$ and $\mathbf{k} = (\Delta k_x, \Delta k_y, k_0 + \Delta k_z)$, yields

$$\frac{\Delta\omega}{\upsilon_g} - \Delta k_z - \frac{1}{2k_0}\left(\Delta k_x^2 + \Delta k_y^2\right) = 0. \tag{7.52}$$

For the envelope $E(x, y, z, t)$ of a linearly polarized pulse propagating in positive z-direction

$$E(x, y, z, t) = \int\int\int d^3\left(\Delta k\right) E(\Delta k_x, \Delta k_y, \Delta k_z) e^{j(\Delta\omega t - \mathbf{\Delta k x})} \tag{7.53}$$

allowing for the nonlinear polarization (from Chapter 3), we obtain

$$\frac{1}{\upsilon_g}\frac{\partial E}{\partial t} + \frac{\partial E}{\partial z} + \frac{j}{2k_0}\nabla_\perp^2 E = -jk_0 \Delta n E. \tag{7.54}$$

In cylindrical coordinates and with the ansatz

$$E(r,z,t) = E_0(r,z,t)\exp\{-j\phi(r,z,t)\}$$

we arrive at the following two equations

$$\left[\frac{1}{v_g}\frac{\partial\phi}{\partial t} + \frac{\partial\phi}{\partial z}\right] + \frac{1}{2k_0}\left[\frac{\partial\phi}{\partial r}\right]^2 = \underbrace{k_0\Delta n} + \underbrace{\frac{1}{2k_0E_0}\left[\frac{\partial^2 E_0}{\partial r^2} + \frac{1}{r}\frac{\partial E_0}{\partial r}\right]} \quad (7.55)$$

$$= \text{self-focusing} + \text{diffraction}$$

$$\left[\frac{1}{v_g}\frac{\partial E_0}{\partial t} + \frac{\partial E_0}{\partial z}\right] + \frac{1}{k_0}\frac{\partial\phi}{\partial r}\frac{\partial E_0}{\partial r} + \frac{1}{2k_0}E_0\left[\frac{\partial^2\phi}{\partial r^2} + \frac{1}{r}\frac{\partial\phi}{\partial r}\right] = 0. \quad (7.56)$$

If a stationary beam exists, for which self-focusing and diffraction exactly balance each other during propagation, then it must hold $\frac{\partial E_0}{\partial t} = \frac{\partial E_0}{\partial z} = 0$, from which in combination with Eq. (7.56) follows

$$\frac{\partial\phi}{\partial r} = 0.$$

I.e., this solution exhibits a plane phase front. Eq. (7.55) then simplifies to

$$-n_2^E E_0^2 = \frac{1}{k_0^2 E_0}\left[\frac{\partial^2 E_0}{\partial r^2} + \frac{1}{r}\frac{\partial E_0}{\partial r}\right]. \quad (7.57)$$

This equation was solved numerically [3, 4]. The stationary solution with the lowest critical power yields

$$P_c = \left(\frac{5.763}{4\pi^2}\right)\frac{\varepsilon_0 c_0 \lambda^2}{n_2^E} = \left(\frac{5.763}{4\pi^2}\right)\frac{\lambda^2}{n n_2^I} \approx \frac{1}{7}\frac{\lambda^2}{n n_2^I}. \tag{7.58}$$

This is of the same order of magnitude as the simple estimate of Eq. (7.49). However, Eq. (7.57) permits to gain deeper insights into the process of self-focusing. It can easily be shown by insertion into Eq. (7.57) that, if $E_0(r)$ is a solution of Eq. (7.57), then also the scaled function $\gamma^2 E_0(\gamma r)$ is a solution. All these solutions contain the same guided energy

$$\int_{-\infty}^{\infty} \gamma^2 E_0^2(\gamma r)\, r dr = \int_{-\infty}^{\infty} E_0^2(r')\, r' dr'$$

$$P = \frac{n_{eff}}{2}\sqrt{\varepsilon_0/\mu_0}\int_{-\infty}^{\infty} E_0^2(r)\, r dr.$$

This scaling invariance is one of the few exact results of self-focusing theory, which reveals that the beam is not stable in 3D. This changes if only one transverse dimension exists, the other dimension could be fixed, e.g., using a waveguide, then it holds according to Eq. (7.54)

$$\frac{1}{v_g}\frac{\partial E}{\partial t} + \frac{\partial E}{\partial z} = -j\frac{1}{2k_0}\frac{\partial^2}{\partial x^2}E - jk_0 n_2^E |E|^2 E. \tag{7.59}$$

Introducing the retarded time $t' = t - z/v_g$, it follows

$$\frac{\partial E(t',z)}{\partial z} = -j\frac{1}{2k_0}\frac{\partial^2}{\partial x^2}E - jk_0 n_2^E \left|E\right|^2 E. \tag{7.60}$$

Again it is straightforward to show by insertion, that this equation, which is called nonlinear Schrödinger equation, possesses solutions

$$E(t',z) = E_0 \mathrm{sech}\left[\frac{x}{x_s}\right] e^{-jk_s z}, \tag{7.61}$$

if the following relations are fulfilled

$$k_s = \frac{1}{2}k_0\frac{n_2^E}{2n}\left|E_0\right|^2, \quad k_s = \frac{1}{2k_0 x_s^2}. \tag{7.62}$$

For a given power density guided in y-direction, that is proportional to

$$\int_{-\infty}^{\infty} E_0^2\left(x\right)dx = 2\left|E_0\right|^2 x_s,$$

there is now only one solution, because the different solutions of form $\gamma^2 E_0^2\left(\gamma x\right)$ belong to different power densities. We will later discuss the properties of the nonlinear Schrödinger equation in greater detail, here we already point out that the solutions (7.61) correspond to a spatial soliton.

For powers far above the critical power for self-focusing, the beam with a Gaussian input profile is focusing down within a distance z_f This distance can be estimated as follows employing a parabolic approximation. The parabolic intensity distribution in the Gaussian beam, $I(r) = I_0 \exp\left[-r^2/w_0^2\right]$, induces in the center of the beam a lens, which bends the phase fronts of the beam

$$\Delta\phi(r) = k_0 n_2^I \left(I(r) - I_0\right) z \approx -k_0 n_2^I I_0 \frac{r^2}{w_0^2} z.$$

This phase shift corresponds to the effect of a lens or a spherical focusing mirror with radius R according to

$$\Delta\phi(r) = -k_0 \frac{r^2}{2R}.$$

As the beam is focusing within a distance $z = z_f \approx R$, it thus follows

$$k_0 n_2^I I_0 \frac{r^2}{w_0^2} z_f = k_0 \frac{r^2}{2z_f}$$

and therefore

$$z_f = \frac{w_0}{\sqrt{2n_2^I I_0}}. \tag{7.63}$$

With the critical power for self-focusing according to Eq. (7.58), we obtain

$$z_f = 0.52 k_0 w_0^2 \sqrt{\frac{P_c}{P}} \approx b \sqrt{\frac{P_c}{P}}. \tag{7.64}$$

Numerical simulations yield

$$z_f = 0.71 b \left(\sqrt{\frac{P}{P_c}} - 0.86 \right)^{-1}. \tag{7.65}$$

As an example, we consider self-focusing in sapphire. At 800-nm wavelength, sapphire has a linear refractive index of about $n = 1.8$ and an intensity-dependent refractive index coefficient of $n_2^I = 3 \times 10^{-16}$ cm^2/W. With this we obtain from Eq. (7.58) a critical power for self-focusing of $P_c = 2.7$ MW.

7.6 Raman and Brillouin scattering

Stimulated Raman and Brillouin scattering is an important technique to investigate oscillations in molecules and solids

They permit the oscillations' identification and study, without them directly coupling to the optical radiation.

stimulated Raman scattering occurring in glass fibers limits the applicable minimum pulse duration in optical communication systems

Raman amplification can be used to realize broadband Raman amplifiers for optical communications.

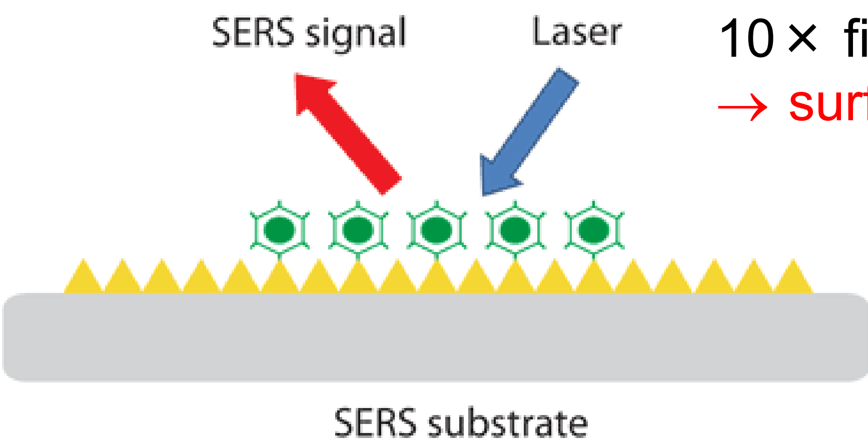
physical effect of Raman scattering:
light propagating though a sample with polarization fluctuations can be scattered in arbitrary direction and shifted in frequency

If the polarization fluctuations originate from oscillations of a molecules or optical or acoustic phonons in a solid, the process takes place via absorption or emission of a phonon, leading to an Anti-Stokes or Stokes shift of the photon absorption or emission of a phonon, leading to an Anti-Stokes or Stokes shift of the photon

$$\omega_{AS} = \omega_L + \Omega \tag{7.66}$$

$$\omega_S = \omega_L - \Omega, \tag{7.67}$$

where ω_L is the frequency of the incident laser photon and Ω the frequency of the phonon involved in the process. A very strongly excited oscillation would contribute equally strongly to Stokes and Anti-Stokes processes. In many cases, the molecule is in the vibrational ground state, thus no thermally populated higher vibrational levels are available. In this case, the Anti-Stokes process is not possible.



from www.semrock.com

10 × field enhancement in nanostructures

→ surface-enhanced Raman scattering (SERS)

A. Campion and P. Kambhampati, Chem. Soc. Rev. **27**, 241 (1998)

For developing a model, we assume that the intramolecular oscillation coordinate Q of the molecule leads to a modulation of the polarizability α at optical frequencies. Then we obtain in linear response

$$\alpha = \alpha_0 + \frac{\partial \alpha}{\partial Q} \cdot Q \tag{7.68}$$

a contribution to the nonlinear polarization of the form

$$P_{NL} = N\varepsilon_0 \frac{\partial \alpha}{\partial Q} QE, \qquad \text{we need } Q \tag{7.69}$$

where we assume, that the electric field itself couples in whatever form to the intramolecular oscillation, N is the density of molecules. One form of this coupling results from the conservation of total energy, i.e., the sum of mechanical and electromagnetic energy. If the total energy is conserved, then the force exerted on the oscillation must be equal to the negative change of the stored electric energy due to the elongation of the oscillation

$$\begin{aligned} F\delta Q &= -\delta\left\{\frac{1}{2}\varepsilon E^2\right\} = -\delta\left\{\frac{1}{2}\varepsilon_0 E^2\left(1+\alpha\right)\right\} \\ &= -\frac{1}{2}\varepsilon_0 E^2 \delta\alpha. \end{aligned} \tag{7.70}$$

with $\varepsilon = \varepsilon_0(1+\alpha)$. This leads to

$$F = -\frac{1}{2}\varepsilon_0 E^2 \frac{\partial \alpha}{\partial Q}. \qquad \propto E^2 \tag{7.71}$$

The oscillation amplitude then satisfies the equation

$$\frac{\partial^2 Q(t,z)}{\partial t^2} + \Gamma \frac{\partial Q(t,z)}{\partial t} + \Omega_0^2 Q(t,z) = \frac{\varepsilon_0}{2m} \frac{\partial \alpha}{\partial Q} E^2(z,t). \tag{7.72}$$

We assume, e.g., that the electric field contains two waves at the laser frequency and the Stokes frequency, i.e.,

$$E(z,t) = E_L e^{j(\omega_L t - k_L z)} + E_S e^{j(\omega_S t - k_S z)} + c.c.$$

Since the resonance frequency of the oscillation Ω_0 is generally far below the optical frequencies ω_L and ω_S, essentially only the difference-frequency terms $\omega_L - \omega_S$ couple to the oscillation

intensity modulation by beat terms

$$\frac{\partial^2 Q(t,z)}{\partial t^2} + \Gamma \frac{\partial Q(t,z)}{\partial t} + \Omega_0^2 Q(t,z) = \frac{\varepsilon_0}{2m} \frac{\partial \alpha}{\partial Q} E_L E_S^* \cdot e^{j\{(\omega_L - \omega_S)t - (k_L - k_S)z\}} + c.c. \tag{7.73}$$

With this we obtain for the stationary oscillation

$$Q(z,t) = \tilde{Q}(z,t) + \tilde{Q}^*(z,t) \tag{7.74}$$

with

$$\tilde{Q}(z,t) = \frac{\frac{\varepsilon_0}{2m} \frac{\partial \alpha}{\partial Q} E_L E_S^*}{\Omega_0^2 - (\omega_L - \omega_S)^2 + j\Gamma(\omega_L - \omega_S)} e^{j\{(\omega_L - \omega_S)t - (k_L - k_S)z\}}. \tag{7.75}$$

Within the Lorentz approximation (i.e., neglecting the off-resonant term, compare Eq. (2.29)), it follows

$$\tilde{Q}(z,t) = \frac{\frac{\varepsilon_0}{4m\Omega_0}\frac{\partial\alpha}{\partial Q}E_L E_S^*}{\Omega_0 - (\omega_L - \omega_S) + j\frac{\Gamma}{2}} e^{\,j\{(\omega_L-\omega_S)t-(k_L-k_S)z\}}. \tag{7.76}$$

For the equation describing the Stokes wave within the SVEA, we then arrive according to Eq. (7.69) at

$$\frac{\partial E_S}{\partial z} = -\frac{j\omega_s}{cn_s}N\frac{\partial\alpha}{\partial Q}\tilde{Q}^* E_L \tag{7.77}$$

all other beat terms ignored here

or

$\chi^{(3)}$

$$\frac{\partial E_S}{\partial z} = \frac{j\omega_S\varepsilon_0 N\left|\frac{\partial\alpha}{\partial Q}\right|^2 |E_L|^2}{4\Omega_0 mcn_s\left\{(\omega_L-\omega_S)-\Omega_0+\frac{j\Gamma}{2}\right\}}E_S. \tag{7.78}$$

The real part of this equation describes gain. With the intensity of the Stokes wave

$$I_S = \frac{n_s}{2}\sqrt{\frac{\varepsilon_0}{\mu_0}}|E_S|^2 \tag{7.79}$$

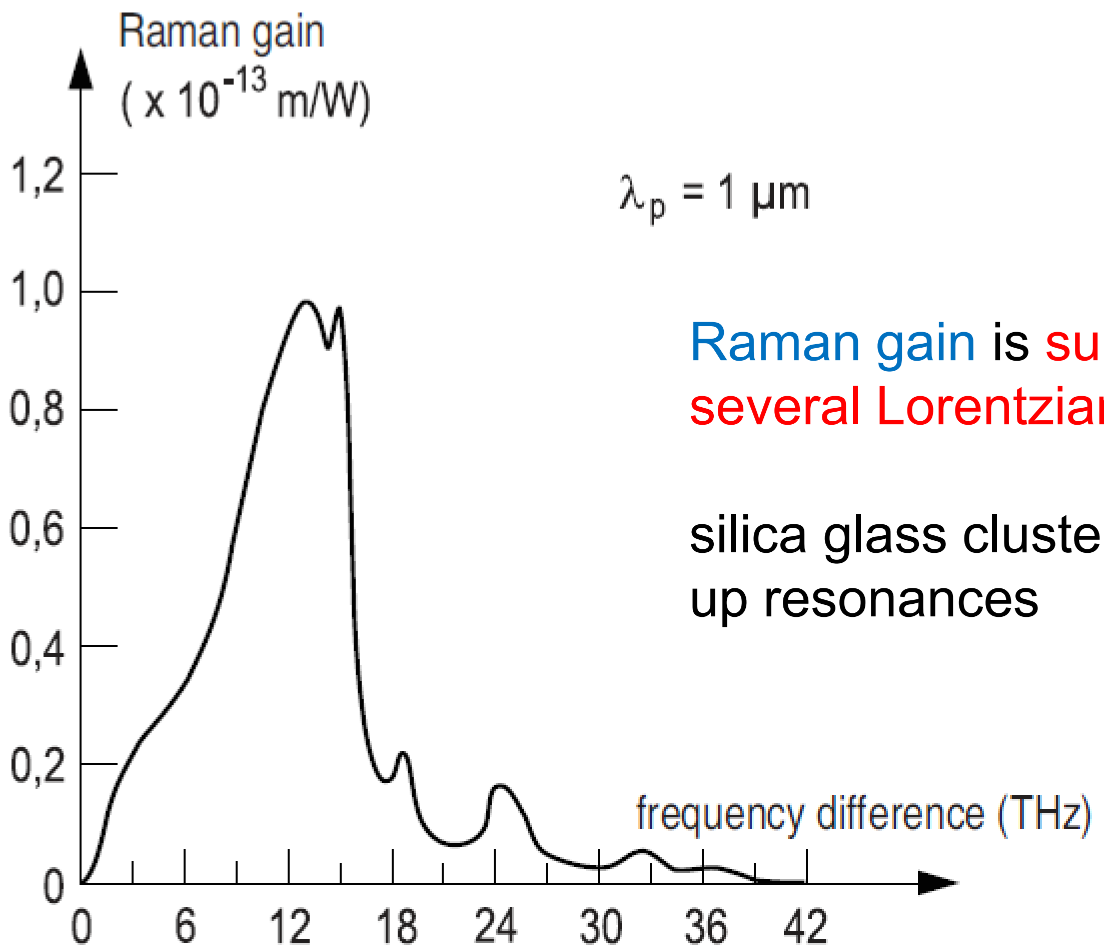
follows

$$I_S(\ell) = I_{S0}\exp\{gI_L\ell\} \tag{7.80}$$

C. V. Raman (Nobel Prize 1930)

with the Raman gain

$$g = \frac{2\omega_s N\left|\frac{\partial\alpha}{\partial Q}\right|^2}{\Omega_0 mc^2 n_s n_L\Gamma}\left\{\frac{\Gamma^2/4}{[\omega_L-\omega_S-\Omega_0]^2+\Gamma^2/4}\right\}. \tag{7.81}$$



Raman gain is superposition of several Lorentzian lines

silica glass clusters that make up resonances

20 nm bandwidth

Figure 7.6: Measured Raman amplification gain of melted quartz at a pump wavelength of 1 μm. The horizontal axis shows difference frequency between laser and Stokes line.

R. H. Stolen, Proc. IEEE **68**, 1232, (1980)