Third Order Nonlinear Processes

P A R T 2

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January 2016

3rd Order Nonlinear Effects (Part 2)

- Self- Phase Modulation
- Generation of Soliton Pulses
- Stimulated Brillouin Scattering (SBS)
- Stimulated Raman Techniques
- Optical Phase Conjugation

Self- Phase Modulation (SPM)

It's the change in the phase of an optical pulse resulting from the nonlinearity of the material refractive index (i.e., Kerr effect).

Due to the Kerr effect, high optical intensity in a medium (e.g. an optical fiber) causes a NL phase delay which has the same temporal shape as the optical intensity. This can be described as a NL change in the refractive index ($\Delta n = n_2 I$)

In the context of self-phase modulation, the emphasis is on the *temporal* dependence of the phase shift, whereas the transverse dependence for some beam profile leads to the phenomenon of self- focusing.

SPM

The third-order nonlinearity gives rise to an intensity-dependent additive to the refractive index:

$$n = n_0 + n_2 I(t) \ ,$$

where n_0 is the refractive index of the medium in the absence of light field, $n_2 = (2\pi/n_0)^2 \chi^{(3)}(\omega; \omega, \omega, -\omega)$ is the nonlinear refractive index, $\chi^{(3)}(\omega; \omega, \omega, -\omega)$ is the third-order nonlinear-optical susceptibility, referred to as the Kerr-type nonlinear susceptibility, and $I(t)$ is the intensity of laser radiation. Then, the nonlinear (intensity-dependent) phase shift of a pulse at a distance L is given by

$$\Phi(t) = \frac{\omega}{c} n_2 I(t) L \ .$$

SPM

Due to the time dependence of the radiation intensity within the light pulse, the nonlinear phase shift is also time-dependent, giving rise to a generally time-dependent frequency deviation:

$$\Delta\omega(t) = \frac{\omega}{c} n_2 L \frac{\partial I}{\partial t} .$$

The resulting spectral broadening of the pulse can be estimated in the following way:

$$\Delta\omega = \frac{\omega}{c} n_2 L \frac{I_0}{\tau}$$

where I_0 is the peak intensity of the light pulse and τ is the pulse duration.

4.5.7 Self-Phase Modulation

If a light pulse of sufficiently high intensity transmits through a material the refractive index becomes a function of the temporally changing intensity of the incident electric light field. The refractive index will be a function of time following the pulse shape of the intensity as a function of time $I_{\mathrm{inc}}(t)$. This temporarily changed refractive index will change the light wavelength in the matter and thus its phase: *self-phase modulation* takes place . As a consequence the frequency of the transmitted light will be tuned during the pulse; it has a *chirp*.

The pulse duration Δt_{FWHM} can be long compared to the reaction time of the matter, which is frequently in the order of a few ps. This may or may not lead to steady-state conditions. In non-stationary self-phase modulation the reaction of the matter will be delayed.

For a simple description a Gaussian pulse shape with the duration $\Delta t_{\mathrm{FWHM}} = 2\Delta\tau\,(\ln 2)^{1/2}$ is assumed:

$$I(t) = I_0 \mathrm{e}^{-(t-t_{\max}/\Delta\tau)^2}. \tag{4.118}$$

During this pulse the refractive index n_{mat} will change under steady-state conditions instantaneously with the intensity (see Fig. 4.29).

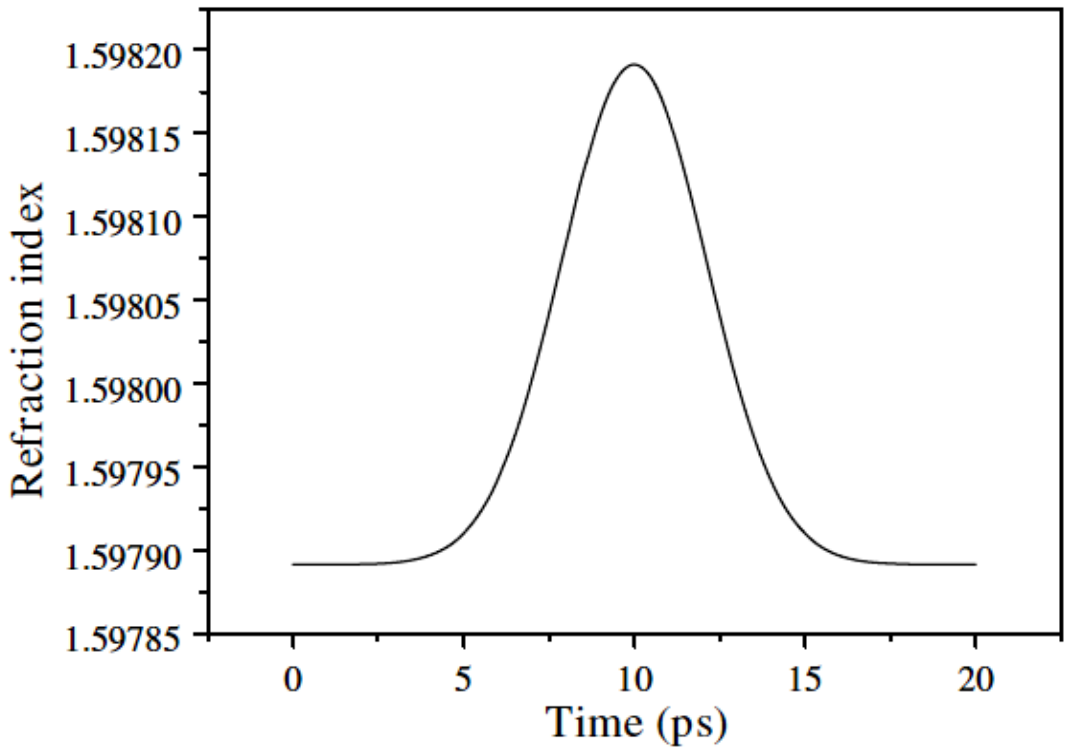


Fig. 4.29. Nonlinear refractive index change under steady state conditions as a function of time for self-phase modulation in CS_2 applying a Gaussian incident pulse with a pulse FWHM duration of 5 ps, the peak at 10 ps, a wavelength of 1064 nm and a maximum power of 10 GW cm^{-2} calculated with the formulas below. In general the change of the refractive index can be delayed if the reaction time of the material is not short compared to the pulse duration

Further almost monochromatic light is presupposed for simplicity which can be achieved experimentally with $1/\Delta\tau \ll \nu_{\mathrm{inc}}$. If the beam is propagating in the z direction through a material with nonlinear refractive index $n_{\mathrm{nl}}(I)$ and length L_{mat} the electric field behind the material is given by:

$$\boldsymbol{E}(t,z) = \boldsymbol{E}_0\, \mathrm{e}^{-\frac{1}{2}(t-t_0/\Delta\tau)^2+\mathrm{i}\varphi} \tag{4.119}$$

with phase factor:

$$\varphi(t, L_{\mathrm{mat}}) = 2\pi\nu_{\mathrm{inc}}t - kL_{\mathrm{mat}} = 2\pi\nu_{\mathrm{inc}}\left(t - \frac{n_{\mathrm{nl}}(I)L_{\mathrm{mat}}}{c_0}\right). \tag{4.120}$$

This phase shift leads to a temporal compression and expansion of the light wave as schematically depicted in Fig. 4.30.

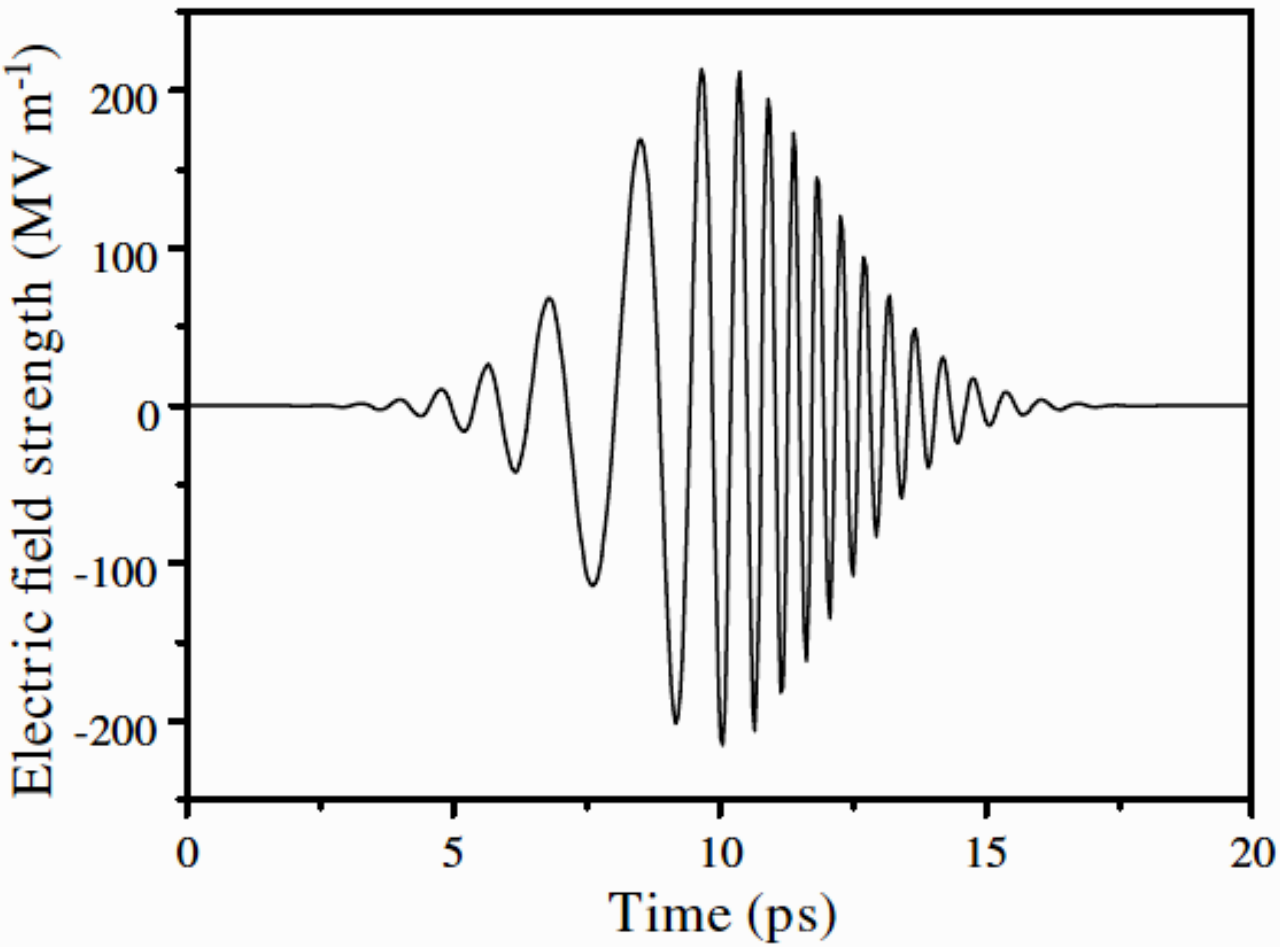


Fig. 4.30. Temporal modulation of the electric field light wave as a consequence of self-phase modulation in a third-order nonlinear material from a short pulse of high intensity (schematic)

Soliton Pulses

Solitons are pulses with a certain balance of nonlinear and dispersive effects.

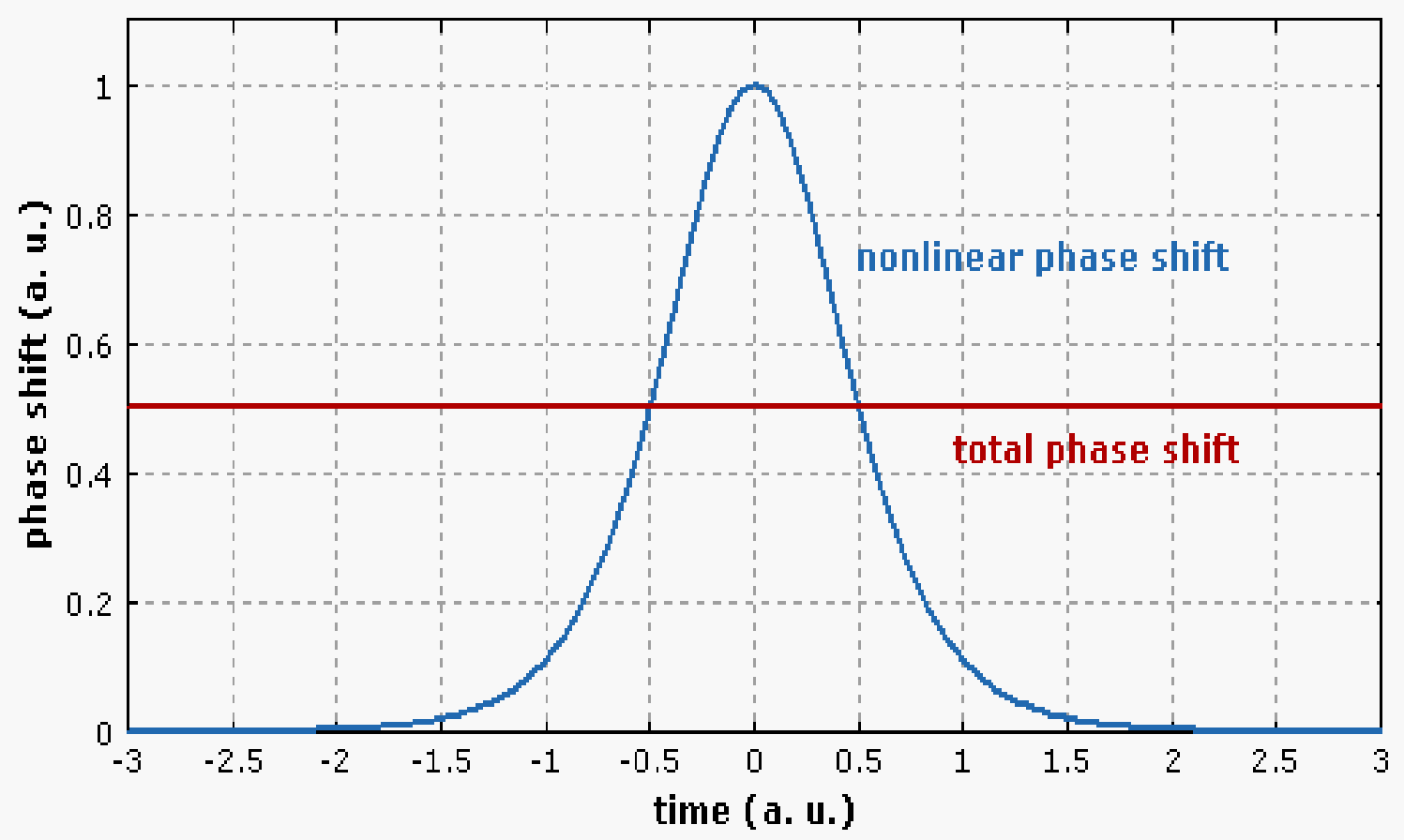
In general, the temporal and spectral shape of a short optical pulse changes during propagation in a transparent medium due to the Kerr effect and chromatic dispersion.

At certain conditions, the effects of Kerr nonlinearity and dispersion can exactly cancel each other, apart from a constant phase delay per unit propagation distance, so that the temporal and spectral shape of the pulses is preserved even over long propagation distances .

The pulse can propagate as a fundamental soliton (or solitary pulse) with constant temporal and spectral shape.

It only acquires a phase shift which the peak of the pulse would experience if only the non-linearity alone were to act on it.

This soliton phase shift is constant over time or frequency, i.e., it does not lead to a chirp or to spectral broadening.



Blue curve time-dependent nonlinear phase shift alone (without dispersion), which is proportional to the optical intensity.

Red curve overall phase shift, resulting from the combined action of nonlinearity and dispersion on a soliton. The constant phase shift does not modify the pulse temporal or spectral shape.

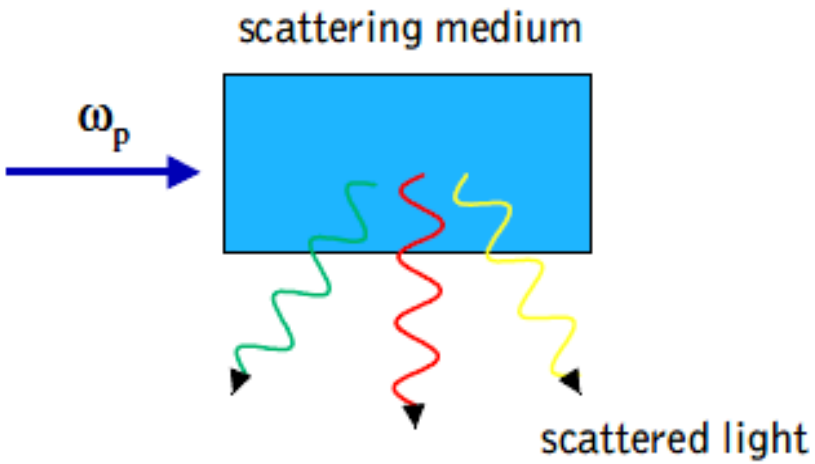
As an example a diode laser pulse of 1.55 μm wavelength and a pulse width of $\Delta t_{\mathrm{FWHM}} = 10\,\mathrm{ps}$ propagates through a silica fiber with a refractive index of 1.45 and $\gamma = 2.8 \cdot 10^{-20}\,\mathrm{m^2\,W^{-1}}$. The soliton period would be about 6 km for a 10 ps pulse and 60 m for a pulse duration of 1 ps and the fundamental soliton needs a peak intensity of $220\,\mathrm{MW\,cm^{-2}}$. For fibers with a diameter of a few μm peak powers in the ten-watt range are necessary for the fundamental soliton.

Solitons are solitary light wave pulses which can superimpose in nonlinear matter without disturbing each other. Thus they can cross or overtake without changing their pulse shape. Only phase changes occur in the interaction.

There are some analogies to the Π-pulses in self-induced transparency but this effect is based on coherent bleaching of the absorption of the material, usually described by a two-level scheme. Therefore the physics of the two processes is completely different.

Solitons in optical fibers can be used for transmitting optical signals over long distances with very high bit rates based on very short pulses. They can also be applied for the generation of short pulses with a duration of a few fs in soliton lasers

Light scattering: A general review

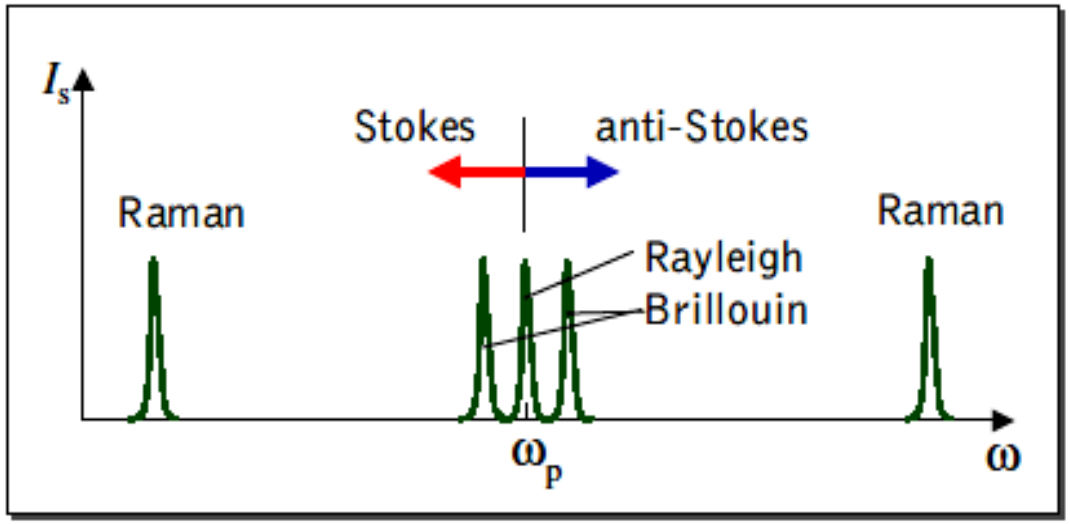


Rayleigh scattering
scattering from *nonpropagating* density fluctuations (elastic)

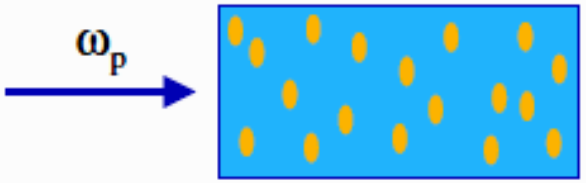
Brillouin scattering
scattering from *propagating* pressure waves (sound waves, acoustic phonons)

Raman scattering
interaction of light with vibrational modes of molecules or lattice vibrations of crystals (scattering from optical phonons)

spectrally resolved detection



Light scattering



Incoming field: $E = E_p \sin \omega_p t$

Induced dipole moment $\vec{\mu} = \alpha \vec{E} = \alpha \vec{E}_p \sin \omega_p t$

Due to molecular vibrations at ω_m

$$\alpha = \alpha_0 + \alpha_1 \sin \omega_m t$$

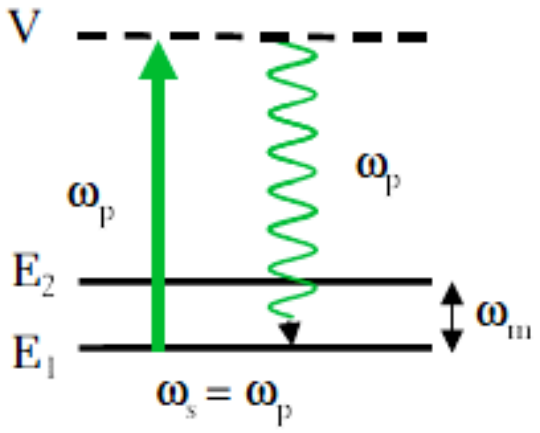
$$\vec{\mu} = \alpha_0 \vec{E}_p \sin \omega_p t + \tfrac{1}{2} \alpha_1 \vec{E}_p \left[\cos(\omega_p - \omega_m)t - \cos(\omega_p + \omega_m)t\right]$$

Induced polarization $P = N\mu$ radiates at ω_p, $\omega_p - \omega_m$, $\omega_p + \omega_m$

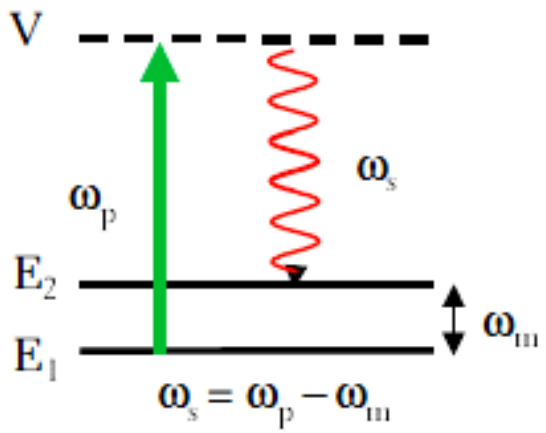
ω_p	Rayleigh scattering (elastic)
$\omega_s = \omega_p - \omega_m$	Stokes Raman scattering (inelastic)
$\omega_{as} = \omega_p - \omega_m$	anti-Stokes Raman scattering (inelastic)

Light scattering

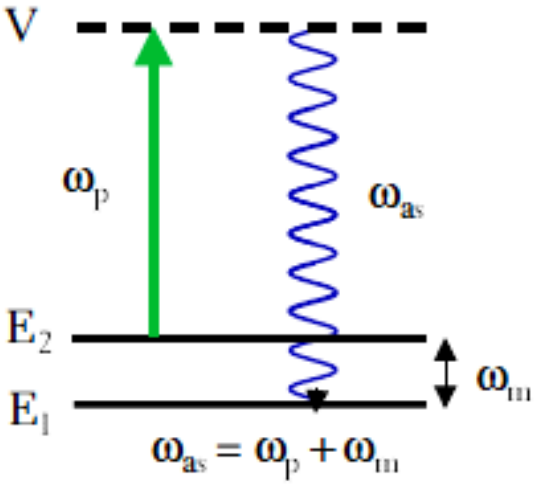
V = virtual level



Stokes



anti-Stokes



Rayleigh scattering

- ✓ parametric process
- ✓ scattered photon into arbitrary direction

Spontaneous Raman scattering

- ✓ spontaneous process
- ✓ scattered photon into arbitrary direction

$$N_2 = N_1 e^{-\hbar\omega_m / kT}$$

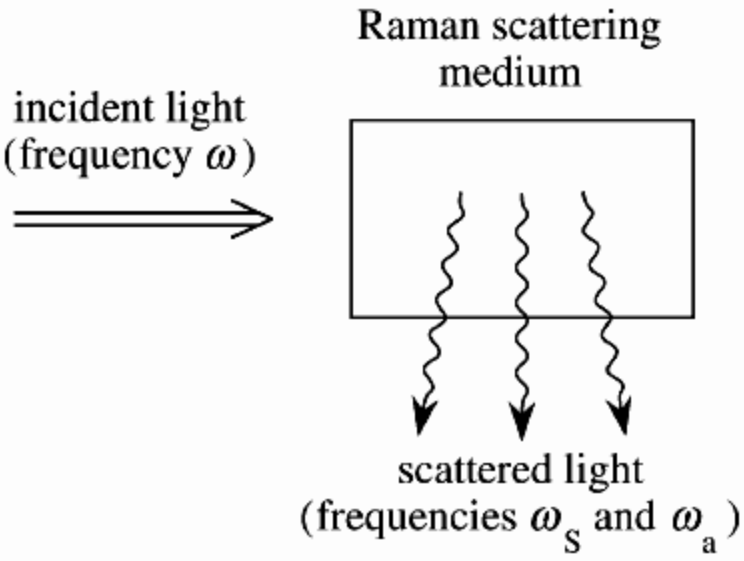
$\Rightarrow$ anti-Stokes lines much weaker

Raman and Brillouin scattering are inelastic scattering processes in which the wavelength of the scattered radiation is different from that of the incident light and a change in the internal energy of the scattering medium occurs. The main distinction between Raman and Brillouin scattering is the type of internal mode involved. Raman scattering involves nonpropagating collective modes in the material. Examples include electronic excitations and molecular vibrations and rotations in liquids and gases, electronic excitations and optical phonons in solids, and electron-plasma oscillations in plasmas. Brillouin scattering involves low-frequency propagating waves, for example acoustic waves in solids, liquids, and gases and ion-acoustic waves in plasmas. The two processes exhibit a range of similarities and differences in the properties of the scattering process as well as in the materials that are involved.

Raman scattering occurs in a wide variety of solids, liquids, gases, and plasmas. The most common form of Raman scattering is one in which the incident light, termed the *pump*, is scattered into light at a longer wavelength, termed the *Stokes wave*, with the energy difference between the incident and scattered photons being taken up in excitation of the appropriate mode of the material. The difference between the incident and scattered photon energy is termed the *Stokes shift*. The identification of the scattered wave as the Stokes wave is made in analogy with the Stokes shift to longer wavelengths in fluorescence, but the dynamics of the two processes are different except for interactions that are resonant with an allowed single-photon resonant transition. Raman scattering in which the incident light is scattered to a light wave at a shorter wavelength, accompanied by a deexcitation of an internal mode of the medium, is termed *anti-Stokes scattering*, and the scattered wave is termed the *anti-Stokes wave*.

Raman scattering

Most of photons scattered from an atom or molecule, are elastically scattered (Rayleigh), i.e., have the same ω and λ as the incident photons. However, a small fraction (1 /10^7) of the scattered photons is inelastically scattered by excitation.



Spontaneous Raman scattering.

To observe this effect, a beam of light illuminates a material sample (which can be a solid, liquid, or gas), and the scattered light is observed spectroscopically, as illustrated in Fig. In general, the scattered light contains frequencies different from those of the excitation source. Those new components shifted to lower frequencies are called Stokes components, and those shifted to higher frequencies are called anti-Stokes components. The Stokes components are typically orders of magnitude more intense than the anti-Stokes components.

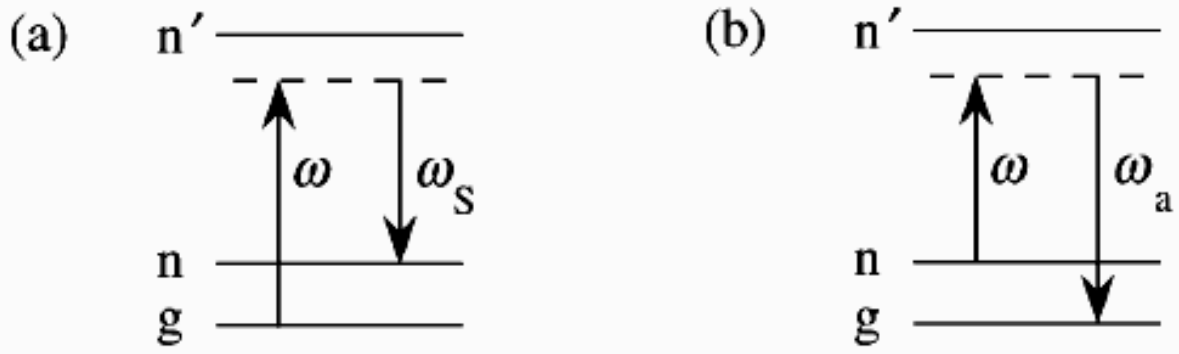
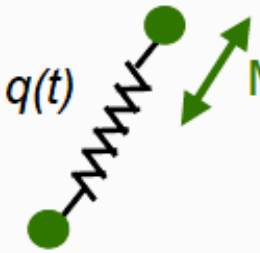


FIGURE Energy level diagrams describing (a) Raman Stokes scattering and (b) Raman anti-Stokes scattering.

These properties of Raman scattering can be understood through use of the energy level diagrams shown in Fig. Raman Stokes scattering consists of a transition from the ground state g to the final state n by means of a virtual intermediate level associated with excited state n'. Raman anti-Stokes scattering entails a transition from level n to level g with n' serving as the intermediate level. The anti-Stokes lines are typically much weaker than the Stokes lines because, in thermal equilibrium, the population of level n is smaller than the population in level g by the Boltzmann factor $\exp(-\hbar\omega_{ng}/kT)$.

Raman Scattering

- ***Spontaneous Raman Scattering - Microscopic origin***



Molecule vibration

$$q - q_0 = q_1 \cos(\omega_v t)$$

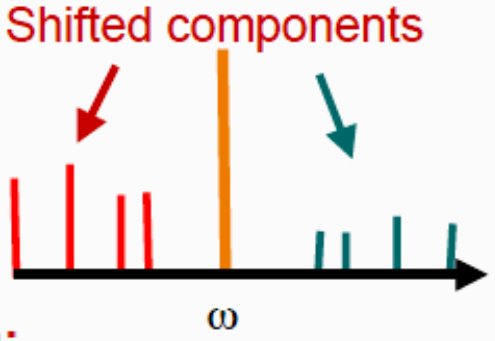
Polarizability of the molecule $\alpha = \alpha_0 + \left(\frac{\partial \alpha}{\partial q}\right)_0 (q - q_0)$

Incident monochromatic wave : $E = E_0 \cos(\omega t)$

Induced dipole : $p = \alpha E$ (α : polarizability of the molecule)

$p(\omega)$ $p(\omega + \omega_v)$ $p(\omega - \omega_v)$

AntiStokes **Stokes**



Origin of the process : polarizability fluctuations

Brillouin Scattering

- ***Origin of the Brillouin scattering***

Inelastic scattering due to the fluctuation of the density of the material

- presence of thermal fluctuations : spontaneous Brillouin scattering
-> scattering of light from acoustic phonons
- density fluctuations reinforced by the beating between two optical waves through electrostriction : stimulated Brillouin scattering

- ***Spontaneous Brillouin scattering***

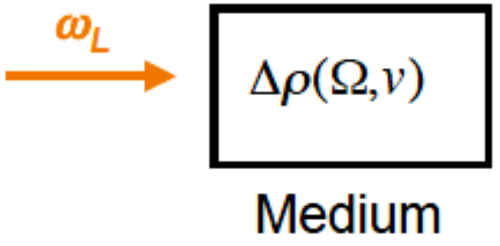
Density fluctuation

$$N = \rho_0 + \Delta\rho\, e^{i(\vec{q}.\vec{r}-\Omega t)} + c.c$$

Average density

Density fluctuation driven by a propagative sound wave

Inelastic scattering onto a sound wave



Brillouin scattering is an interaction between EM wave and a density wave (**photon-phonon scattering**)

The scattering is **inelastic**: the photon may lose (Stokes process) or gain energy (anti-Stokes process). This shift in photon frequency, known as the **Brillouin shift**, is equal to the energy of the interacting phonon.

when the medium is compressed its index of refraction changes, and a fraction of the traveling light wave, interacting with the periodic refraction index variations, is deflected as in 3D diffraction grating. Since the sound wave, too, is travelling, light is also subjected to a **Doppler shift**, so its frequency changes.

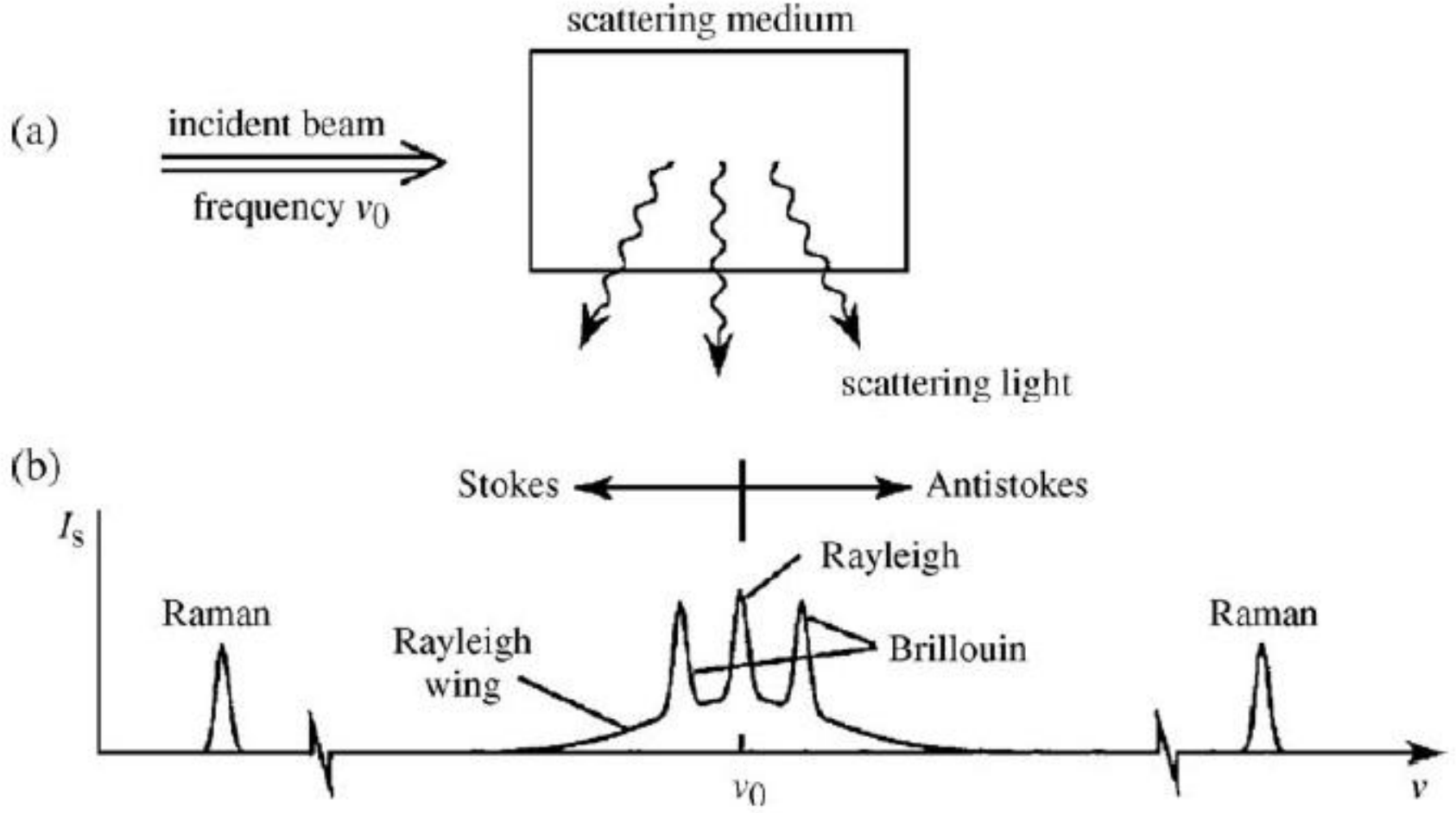


FIGURE 8.1.1 Spontaneous light scattering. (a) Experimental setup. (b) Typical observed spectrum.

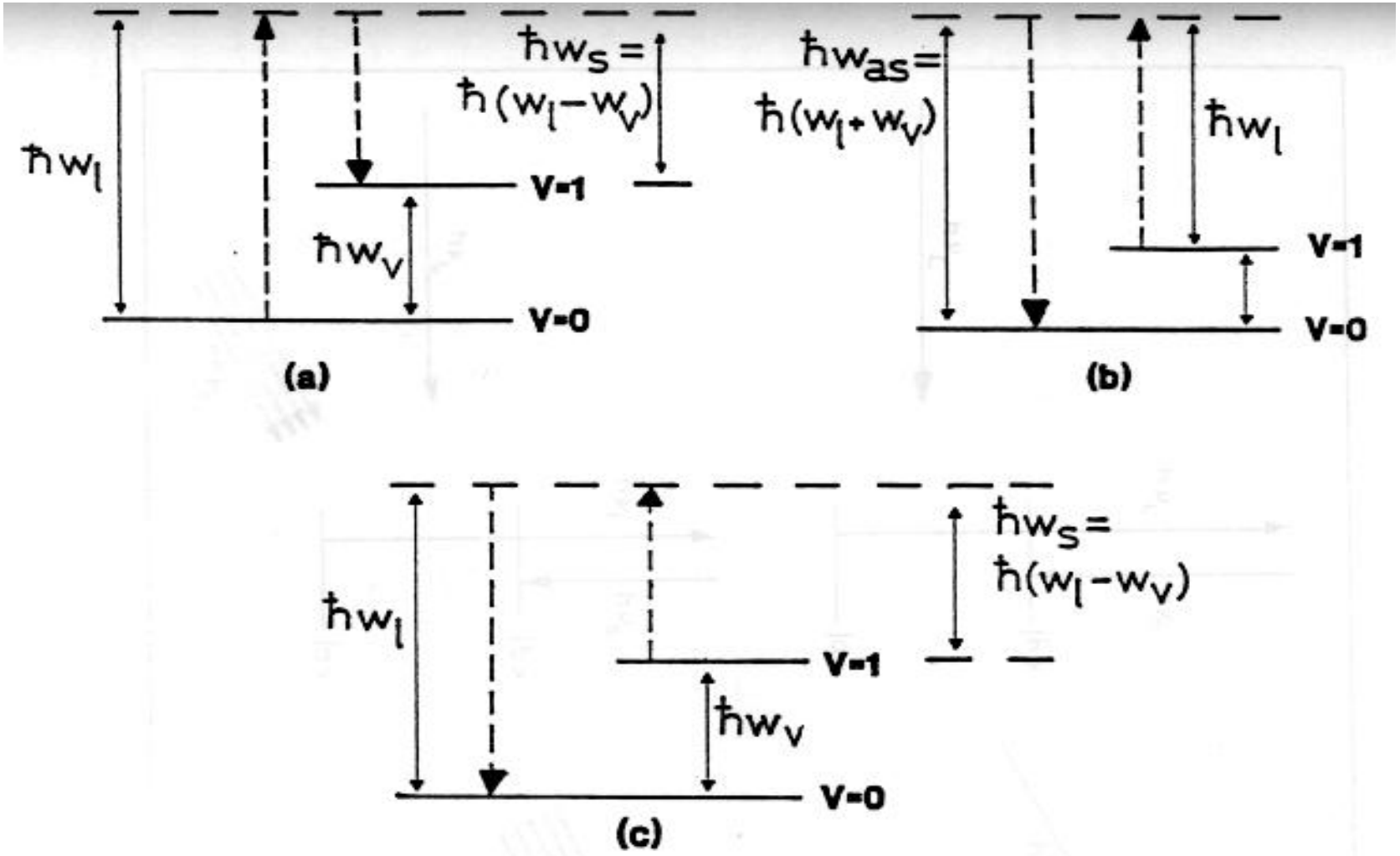
Stimulated Scattering

Light can scatter inelastically from fundamental excitations in the medium, resulting in the production of light at a frequency that is shifted from that of the incident by the excitation frequency.

e.g., Brillouin scattering from acoustic vibrations; various forms of Raman scattering involving molecular rotations or vibrations, electronic states in atoms or molecules, lattice vibrations.

At high power levels, the scattered light experiences exponential **gain**, and the process is then termed **stimulated**.

In stimulated scattering, the energy of the incident light is almost completely converted to another, scattered wave at lower ω (longer λ) with the small energy difference being release as phonons.



(a) A Stokes scattering in which a laser photon at ω_l is absorbed, while a Stokes $(\omega_l - \omega_v)$ photon is created along with a vibrational (v = 1) quantum. (b) An anti-Stokes scattering in which a laser photon at ω_l and a vibrational (ω_v) quantum are absorbed, while a photon at $\omega_l + \omega_v$ is created. (c) A process in which the presence of laser radiation at ω_l stimulates the absorption of Stokes photons at $\omega_l - \omega_v$, that is, the reverse of (a).

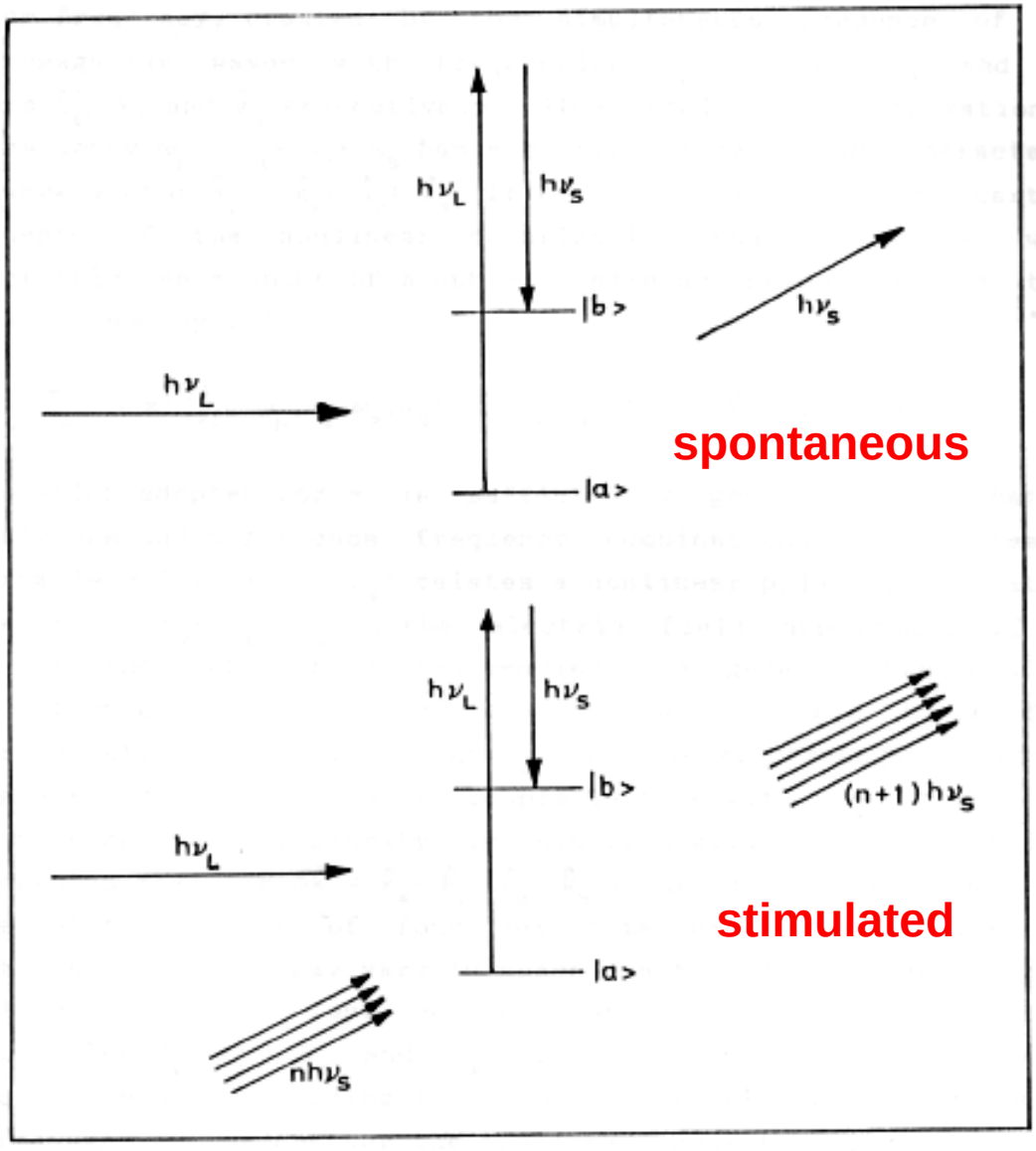


Diagram of spontaneous (top) and stimulated (bottom) Raman scattering as a quantum process.

The nonlinearity of the dielectric medium provides a coupling between different homogeneous plane waves. Consider, for example, the existence of a polarization at the sum frequency, created by the simultaneous presence of three electromagnetic waves with frequencies ω_1, ω_2 and ω_3 and wave vectors $\bar{k}_1$, $\bar{k}_2$ and $\bar{k}_3$ respectively. This nonlinear polarization at the frequency $\omega_p = \omega_1 + \omega_2 + \omega_3$ has a spatial distribution characterized by a wave vector $\bar{k}_p = \vec{k}_1 + \vec{k}_2 + \vec{k}_3$. If i, j, k and l define cartesian components of the nonlinear polarization and the three waves, respectively, an element of a cubic nonlinear susceptibility tensor may be defined by

$$\vec{P}_i^{NL}(\omega_p, \vec{k}_p) = \chi^{(3)}_{ijkl}(-\omega_p, \omega_1, \omega_2, \omega_3)\, \vec{E}_j(\omega_1, \vec{k}_1)\, \vec{E}_k(\omega_2, \vec{k}_2)\, \vec{E}_l(\omega_3, \vec{k}_3)$$

Stimulated Brillouin Scattering (SBS)

Stimulated Brillouin scattering (SBS) is an important third-order nonlinear optical effect that has been widely used for efficient phase conjugate reflection of high-power lasers.[26] An incident laser beam can scatter with the periodic refractive index variations associated with a propagating acoustic wave. The scattered light, depending on the propagation direction of the acoustic wave, will be Stokes or anti-Stokes shifted by the frequency of the acoustic wave. The process is stimulated because the interference of the incident and scattered wave can lead to an amplification of the acoustic wave, which then tends to pump more energy into the scattered wave. This positive feedback process can cause an exponential growth of the SBS beam and very high efficiencies in the right circumstances. Optical feedback to the medium is accomplished in one of two ways: (1) *electrostriction* is local compression of the material in response to the strength of the electromagnetic field with a commensurate refractive index change; and (2) *linear optical absorption* by the laser field leads to local heating, expansion, density fluctuations, and thus periodic modulation of the refractive index. The latter effect is an example of a cascaded $\chi^{(1)}$:$\chi^{(1)}$ process

Brillouin scattering involves low-frequency propagating waves—for example, acoustic waves in solids, liquids, and gases and ion-acoustic waves in plasmas. Again, scattering can be to a longer or shorter wavelength than the incident radiation, with the long-wavelength scattered wave being termed the *Stokes wave* and the short-wavelength scattered wave termed the *anti-Stokes* wave. The difference between the incident and scattered frequencies is again termed the *Stokes shift* or *anti-Stokes shift* as appropriate. For Brillouin scattering, the energies of the modes are much lower than for Raman scattering, and anti-Stokes radiation is much more common. Common Brillouin shifts are typically on the order of 0.1 to 100 GHz, and depend on the excitation wavelength and interaction geometry as well as on material properties. Brillouin scattering is used most commonly for phase conjugation and pulse compression. It is also prominent as a limiting process for intensity in fiber-optic systems.

The frequency of the reflected beam is slightly lower than that of the incident beam; the frequency difference ν_B corresponds to the frequency of emitted phonons. This so-called Brillouin frequency shift is set by a phase-matching requirement. For pure backward Brillouin scattering, the Brillouin shift can be calculated from the refractive index n, the acoustic velocity v_a and the vacuum wavelength λ :

$$\nu_B = \frac{2nv_a}{\lambda}$$

The Brillouin frequency shift depends on the material composition and to some extent the temperature and pressure of the medium.

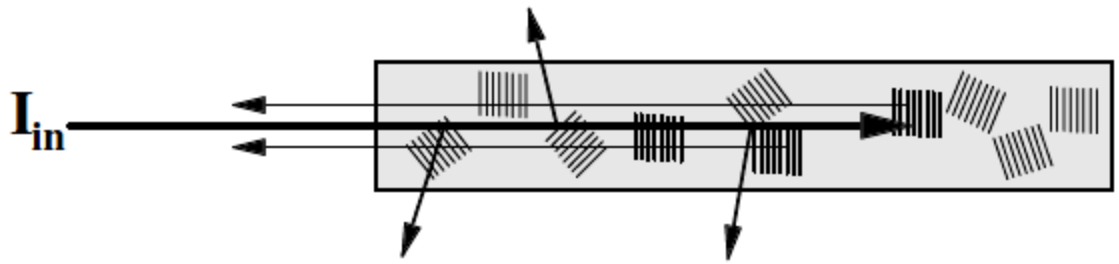
4.5.9 Stimulated Brillouin Scattering (SBS)

Stimulated Brillouin scattering (SBS) occurs as amplified spontaneous Brillouin scattering (see Sect. 3.11.3, p. 163) at sufficiently high intensities of the incident light wave, which increases the sound wave amplitude by electrostriction. It occurs also as spontaneous scattering, in the nonresonant spectral range. But SBS is an inelastic optical scattering process with very small energy loss, as it excites an acoustic phonon of the hyper-sound wave.

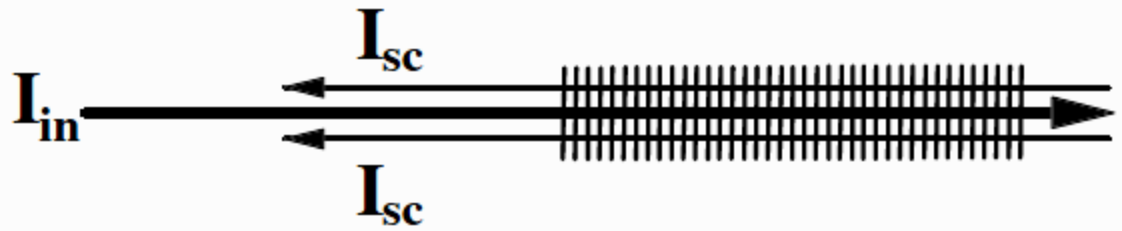
SBS is applied in phase conjugating mirrors (PCM) (see Sect. 4.5.14, p. 250), e.g. for the improvement of the beam quality of solid-state lasers

Although SBS is a $\chi^{(3)}$ process the phase conjugation of these SBS mirrors can be made by simply focusing only one light beam in a cell with a suitable gas or liquid or in a solid SBS-material.

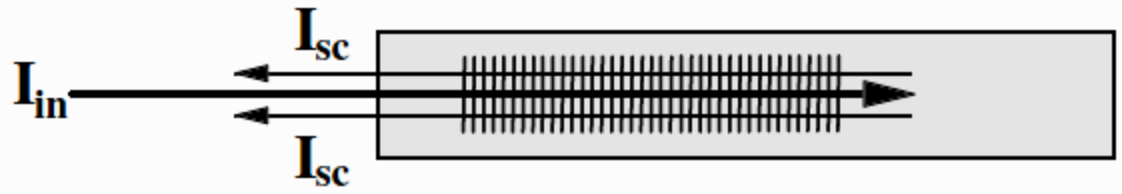
The third-order nonlinear process can be imagined in four steps which take place simultaneously:



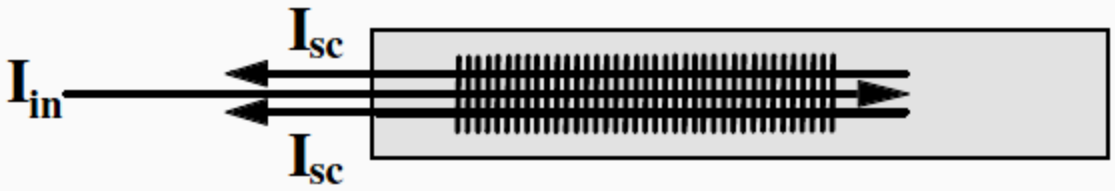
(i) a small share of the spontaneous scattered light will exactly be back-reflected towards the incident beam (scattered light is slightly frequency shifted);



(ii) incident and reflected light waves interfere and generate a moving intensity grating;



(iii) this intensity grating amplifies the suitable sound wave for back-scattering via electrostriction;



(iv) the amplified sound wave increases the share of the exactly back-scattered light which leads to more interference and the process continues at (i) with positive feedback up to saturation (reflectivity up to 100%)

Because of the perfect back-scattering in SBS the scattering angle is 180° and thus the wavelength of the sound wave Λ_{sound} results from the wavelength of the incident light wave λ_{inc}:

$$\text{sound wavelength} \quad \Lambda_{\text{sound}} = \frac{1}{2}\lambda_{\text{inc}}. \tag{4.131}$$

The sound wave frequency Ω_{sound} follows from the sound velocity v_{sound}:

$$\text{sound frequency} \quad \Omega_{\text{sound}} = 2\frac{v_{\text{sound}}}{\lambda_{\text{inc}}} = 2\frac{v_{\text{sound}} n \nu_{\text{inc}}}{c_0} \tag{4.132}$$

and is in the range of several 100 MHz for gases and up to several 10 GHz for liquids and solids (see Table 4.8, p. 228). Thus the energy loss of the light in the range 10^{-4}–10^{-6} can often be neglected but it will be important if interference of the original pump beam and the reflected light is to be used in applications.

The potential linewidth of the SBS $\Delta\Omega_{\text{sound}}$ measured at 1/e of the peak value is a function of the lifetime of the sound wave τ_{sound}:

$$\text{SBS linewidth} \quad \Delta\Omega_{\text{sound}} = \frac{1}{2\pi\tau_{\text{sound}}} \tag{4.133}$$

which is twice the lifetime of the phonons τ_{SBS}:

$$\text{phonon lifetime} \quad \tau_{\text{SBS}} = \frac{1}{2}\tau_{\text{sound}} = \frac{K_{\text{sound}}^2\eta}{\rho_0} \propto \frac{1}{\lambda_{\text{inc}}^2} \tag{4.134}$$

with the material viscosity η. The reciprocal quadratic wavelength dependency is a rough approximation quite well fulfilled in the near UV to IR range.

$$\textbf{stationary Brillouin gain} \quad g_{\text{SBS}} = \frac{(2\pi\Omega_{\text{sound}}\gamma^{\text{e}})^2\tau_{\text{sound}}}{c_0^3 n\rho_{\text{mat},0}v_{\text{sound}}} \tag{4.147}$$

The intensity of the reflected light I_{scatt} can be written as:

$$I_{\text{scatt}}(L_{\text{interaction}}) = I_{\text{scatt,spont}}(z_0)\, \mathrm{e}^{g_{\text{SBS}} I_{\text{inc}} L_{\text{interaction}}} \tag{4.152}$$

with the interaction length $L_{\text{interaction}}$ and assuming the incident beam and the reflected beam interfere coherently. The total stationary SBS gain G_{SBS} results from:

$$\textbf{stationary SBS-gain} \quad G_{\text{SBS}} = g_{\text{SBS}} I_{\text{inc}} L_{\text{interaction}}. \tag{4.153}$$

The stationary "SBS-threshold" power P_{th} can be estimated from this formula by considering the spontaneous reflectivity useful for starting the SBS. It is in the range of $R_{\text{spontaneous}} = 10^{-11}$ and thus the total SBS gain G_{SBS} has to be bigger than approximately 20 for 2% reflectivity:

$$\text{stationary SBS-threshold} \quad P_{\text{th}} \approx 20 \frac{A_{\text{interaction}}}{g_{\text{SBS}} L_{\text{interaction}}}. \tag{4.154}$$

Typical cross-sections $A_{\text{interaction}}$ in SBS with focused beams are of the order of $10^{-5}\,\text{cm}^2$ and the interaction lengths are a few mm. Thus gases and solids show thresholds of several 100 kW to MW and liquids can have values as low as 10 kW

Stimulated Raman Scattering (SRS)

Introduction

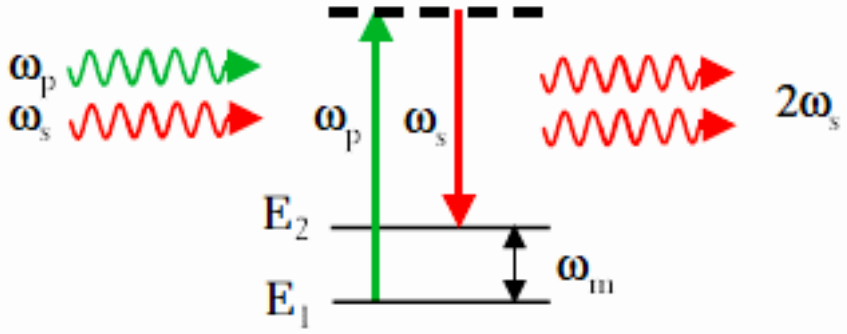
Raman scattering can occur in the spontaneous and stimulated regimes. In the spontaneous regime, the power of the Stokes and anti-Stokes waves is proportional to the power of the pump wave. The entire manifold of Raman-active internal modes is present in the scattered spectrum, with relative intensities of the Stokes components being determined by the relative Raman scattering cross sections for the various modes. Anti-Stokes scattering arises in spontaneous Raman scattering through thermal excitation of the internal modes. Therefore, the intensity of anti-Stokes modes is reduced from that of the Stokes modes for the same internal level by the thermal excitation factor $e^{-\hbar\omega_o/kT}$. Anti-Stokes scattering in the spontaneous regime is generally less common than Stokes scattering, except when low-lying rotational levels of molecules or low-frequency phonons in solids are involved, because thermal excitation of the internal mode is required.

Stimulated Raman Scattering (SRS) occurs when the intensity of the incident pump wave is strong enough to initiate a positive feedback effect in the medium, resulting in exponential growth of the scattered wave. Stimulated Raman scattering is used for wavelength shifting of coherent light, amplification, improved optical beam properties, pulse compression, phase conjugation, and beam combining.

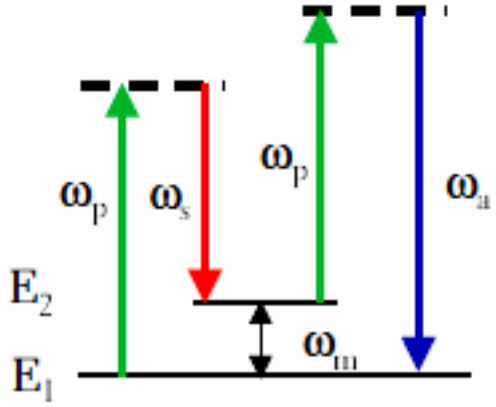
An instantaneous process, in which a Stokes photon stimulates the emission of another Stokes photon. Anti-Stokes photons result from four-wave mixing.

Stimulated Raman scattering

For strong pump fields the number of emitted Raman photons is high
⇒ they can stimulate further (coherent) Raman photons to be emitted in the direction of the pump beam
⇒ gain !



generation of stimulated Stokes radiation



Stokes - anti-Stokes coupling

$$\hbar\omega_p + \hbar\omega_s = 2\hbar\omega_s + \hbar\omega_m$$

⇒ Stokes and anti-Stokes waves can have comparable intensities

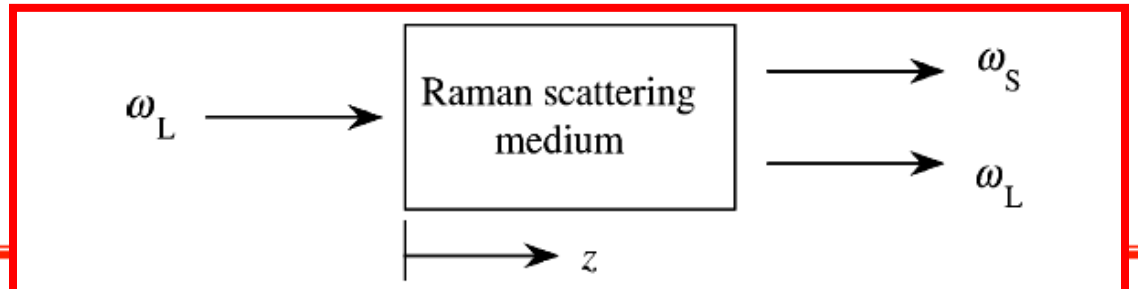
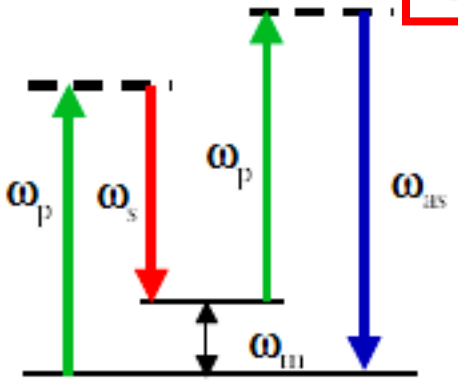


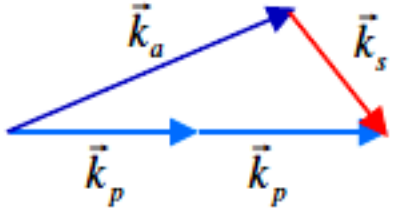
FIGURE 10.2.1 Geometry describing stimulated Raman scattering.



Parametric process

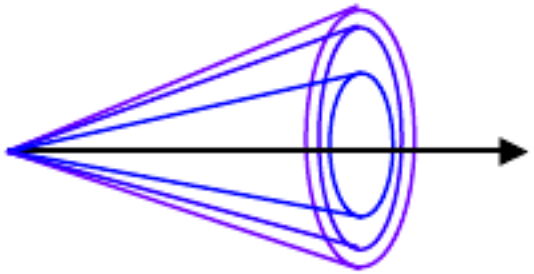
Energy conservation: $2\hbar\omega_p = \hbar\omega_s + \hbar\omega_{as}$

Momentum conservation: $2\vec{k}_p = \vec{k}_s + \vec{k}_{as}$



In gases: $n(\omega_p) \approx n(\omega_s) \approx n(\omega_{as}) \approx 1$

$\Rightarrow \sim$ collinear geometry



anti-Stokes radiation cones for different ω_m

Stimulated Raman scattering

- ✓ Threshold pump intensity
- ✓ $\sigma_{stim} \approx 10^5 \times \sigma_{spont}$
  - ✓ high conversion efficiency (> 10 %), but requires a strong pump
- ✓ Stokes and anti-Stokes lines have comparable intensities
- ✓ Applications: Fiber Raman laser, Raman shifter, spectroscopy

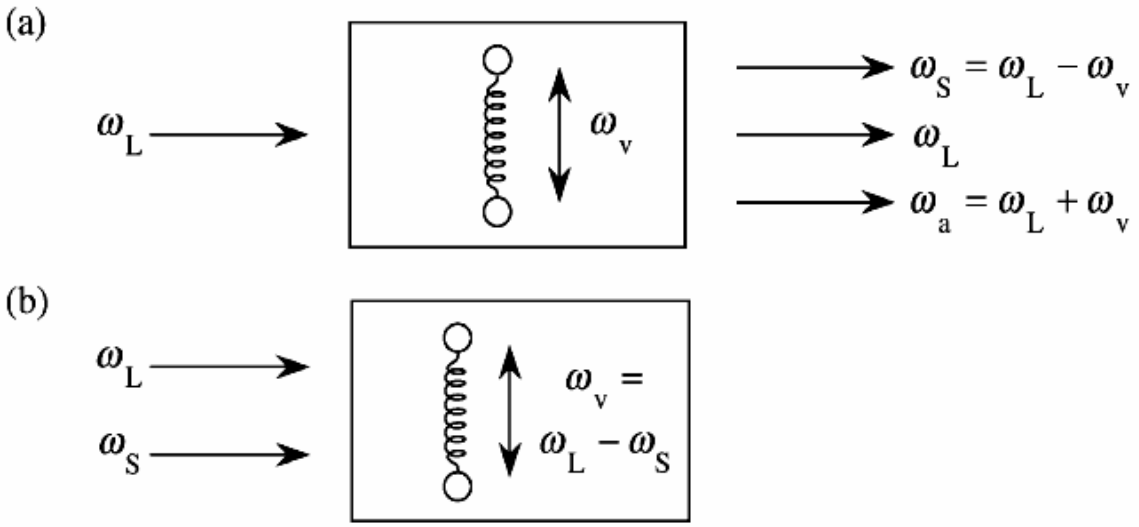


FIGURE Stimulated Raman scattering.

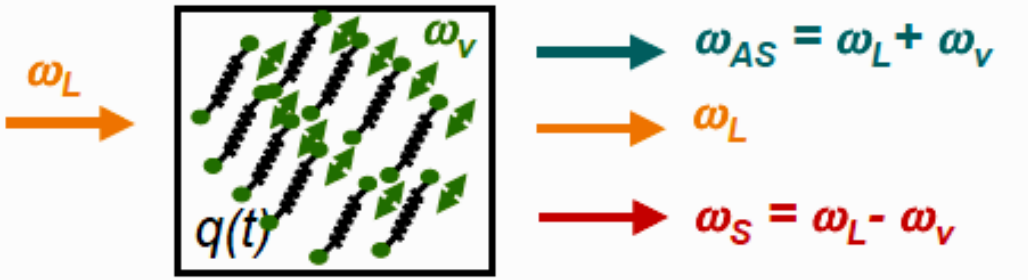
The origin of stimulated Raman scattering can be understood schematically in terms of the interactions shown in Fig. . Part (a) of the figure shows how molecular vibrations modulate the refractive index of the medium at frequency ω_v and thereby impress frequency sidebands onto the laser field. Part (b) shows how the Stokes field at frequency $\omega_S = \omega_L - \omega_v$ can beat with the laser field to produce a modulation of the total intensity of the form

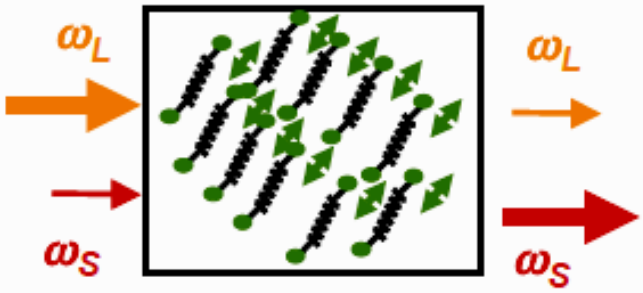
$$\tilde{I}(t) = I_0 + I_1 \cos(\omega_L - \omega_S)t.$$

This modulated intensity coherently excites the molecular oscillation at frequency $\omega_L - \omega_S = \omega_v$. The two processes shown in parts (a) and (b) of the figure reinforce one another in the sense that the interaction shown in part (b) leads to a stronger molecular vibration

- ***Stimulated Raman scattering - Classical behavior***

Molecules in vibration





- The interference between the wave components ω_L ***and*** ω_S maintains, strengthens the molecule vibration @ ω_L
- Consequence : amplification of the signal @ ω_S

Energy required to drive the dipole oscillation : $W = \frac{1}{2}\langle \mathbf{p}(z,t).\mathbf{E}(z,t) \rangle$

$$\mathbf{p} = \alpha \mathbf{E} \quad \Rightarrow \quad W = \frac{1}{2}\alpha \langle \mathbf{E}^2(z,t) \rangle$$

Time average

- ***Stimulated Raman scattering - Classical behavior***

$$P_{NL}(\omega_S) = 6\epsilon_0 \chi_R^{(3)}(\omega_S; \omega_L, -\omega_L, \omega_S)|A_L|^2 A_S e^{ikz}$$

Degeneracy factor

$$\chi_R^{(3)}(\omega_S) = \frac{6\,\frac{N}{\epsilon_0 m}\left(\frac{d\alpha}{dq}\right)^2}{\omega_v^2 - (\omega_L - \omega_S)^2 + 2i(\omega_L - \omega_S)\gamma}$$

Susceptibility expression shows that in a resonance case i.e. $\omega_v = \omega_L - \omega_S$

$\chi_R^{(3)}(\omega_S) =$ negative imaginary

- ***Stimulated Raman scattering***

Nonlinear polarization calculation @ ω_S (ω_{AS}) et ω_L solving the equation of motion of a classical harmonic oscillator :

$$P_{NL}(\omega_S) = 6\varepsilon_0 \chi_R^{(3)}(\omega_S;\omega_L,-\omega_L,\omega_S)|A_L|^2 A_S e^{ik_S z}$$

$$P_{NL}(\omega_L) = 6\varepsilon_0 \chi_R^{(3)}(\omega_L;\omega_S,-\omega_S,\omega_L)|A_S|^2 A_L e^{ik_L z}$$

Susceptibility expression shows that in a resonance case i.e. $\omega_v = \omega_L - \omega_S$

$$\chi_R^{(3)}(\omega_S) = \chi_R^{(3)}(\omega_L)^* = \quad \text{negative imaginary}$$

Coupled equations :

$$\begin{cases} \dfrac{\partial A_S}{\partial z} = g_R |A_L|^2 A_S \\ \dfrac{\partial A_L}{\partial z} = -\dfrac{\omega_L}{\omega_S} g_R |A_S|^2 A_L \end{cases}$$

Stokes wave amplification

Pump wave depletion

with $g_R = \dfrac{3\omega_S}{2nc}\chi_R^{(3)}(\omega_S)$

Dispersion relation for sound waves

$$\Omega = v\,|q| \qquad \text{with } v \text{ : sound velocity within the medium}$$

Induced macroscopic polarization due to density fluctuations of the medium

$$\vec{P}_L = N(0,\Omega)\;\alpha\;\vec{E}_L(\omega_L)$$

$$P(\omega_L)$$

$$P(\omega_S = \omega_L - \Omega) \qquad \text{Stokes wave}$$

$$P(\omega_{AS} = \omega_L + \Omega) \qquad \text{Anti-Stokes wave}$$

Phase matching condition

$$\vec{k}_S = \vec{k}_L - \vec{q}$$

$$\vec{k}_{AS} = \vec{k}_L + \vec{q}$$

4.5.13 Stimulated Raman Techniques

Although stimulated Raman scattering (SRS) is usually applied in the nonabsorbing spectral range of matter the energy change of the scattered photons compared to the energy of the incident photons can be as large as 10%. As in spontaneous Raman scattering the SRS process can be understood as scattering coupled with a matter transition between two vibrational energy states with energy difference $E_{\mathrm{vib}} = h\nu_{\mathrm{vib}}$ via a virtual energy state of more than ten times this energy. Different measuring techniques are used as will

4.5.13.1 Stimulated Raman Scattering (SRS)

Stimulated Raman scattering (SRS) can take place with the excitation of the vibration or of an optical phonon (Stokes SRS results in smaller photon energy) or by its depletion (anti-Stokes SRS):

energy condition

$$\nu_{\text{scatt,SRS}} = \nu_{\text{inc}} \mp m\nu_{\text{vib}} \quad \text{with} \quad m = 1, 2, 3, \ldots \tag{4.182}$$

whereas subsequent scattering or nonlinear coupling of molecular vibration allows multiple frequency shifts with $m > 1$. The vibrational overtones may show slightly shifted frequencies ν_{vib} due to the anharmonicity of the vibrational potential. Again, as in spontaneous Raman scattering the selection rules demand polarizability for the considered vibration in contrast to the necessity of a dipole moment of this vibration to detect it in IR spectroscopy. Rotational transition and translation energies may overlay the spectra in both cases and produce complicated structures or broadening. This allows detailed analysis of the matter structure regarding the bond lengths, potentials and masses of the atoms involved if high-resolution techniques are applied.

The intensity of the scattered light in SRS can be amplified by many orders of magnitude from spontaneous Raman scattering up to several 10% of the intensity of the incident light. Under certain circumstances strength of the anti-Stokes SRS can be comparable to the Stokes SRS. It occurs in small cones, of a few degrees in the forward and backward direction.

4.5.13.2 Inverse Raman Spectroscopy (IRS)

Inverse Raman spectroscopy (IRS) is obtained if the depletion of a weak probe light signal in the linear intensity range at the frequency of the SRS pump transition is measured while a strong laser is tuned across the Stokes frequency of the matter as schematically shown in Fig. 4.42 [M15].

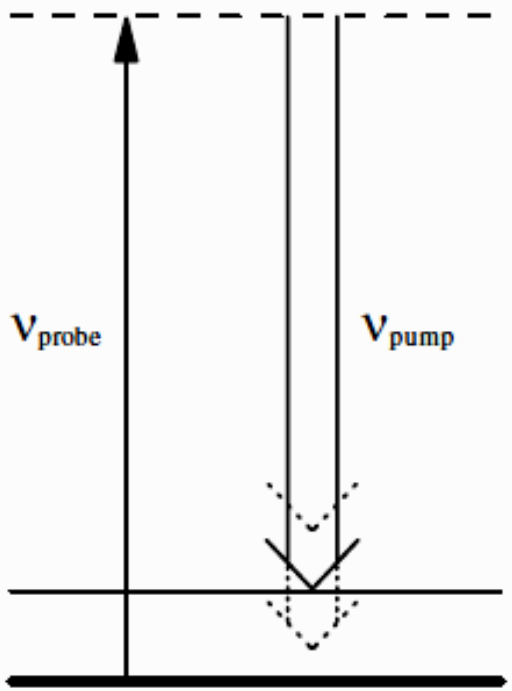


Fig. 4.42. Inverse Raman spectroscopy (IRS) with measurement of the depletion of a weak probe signal at the frequency of the SRS pump as a function of the frequency of a strong and tunable laser at the Stokes wavelength

4.5.13.4 Coherent Anti-Stokes Raman Scattering (CARS)

The combination of simultaneous stimulated Stokes and anti-Stokes Raman scattering leads to the interaction of four photons in the matter [4.593–4.623]. In coherent anti-Stokes Raman scattering (CARS) two strong laser beams with frequencies ν_{inc} and $\nu_{\mathrm{SRS,S}}$ are applied (see Fig. 4.44).

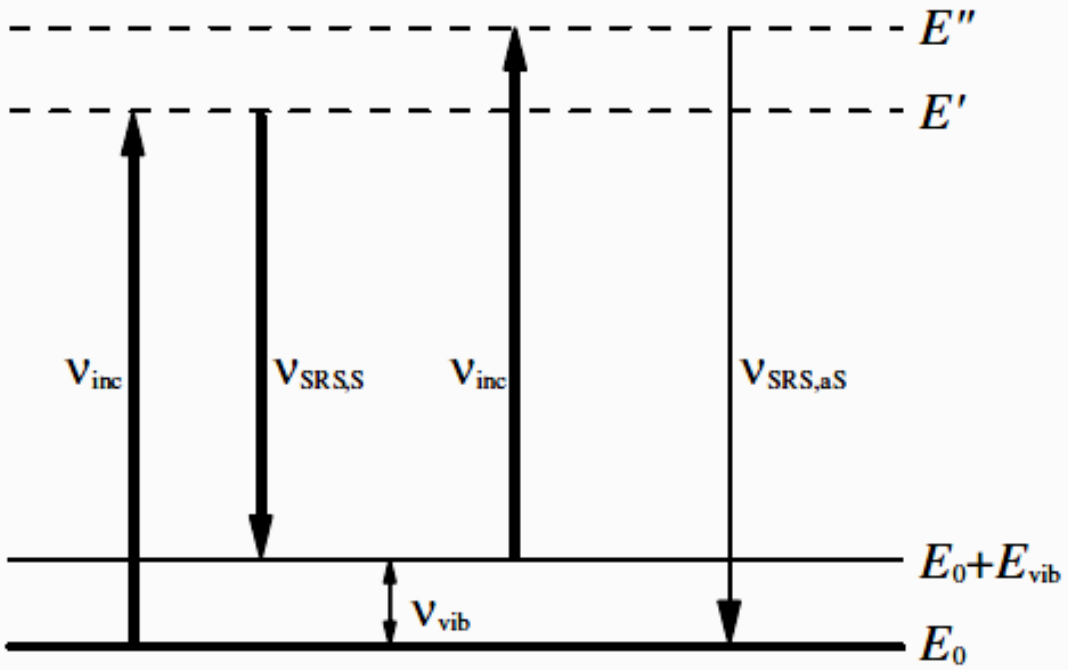


Fig. 4.44. Coherent anti-Stokes Raman scattering (CARS) pumping with two laser beams with frequencies ν_{inc} and $\nu_{\mathrm{SRS,S}}$, obtaining the anti-Stokes Raman light with $\nu_{\mathrm{SRS,aS}}$. For strong signals phase matching has to be achieved

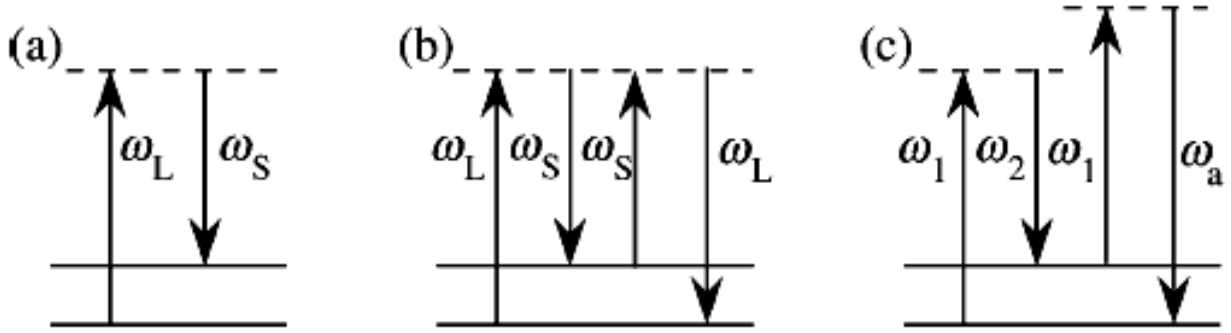


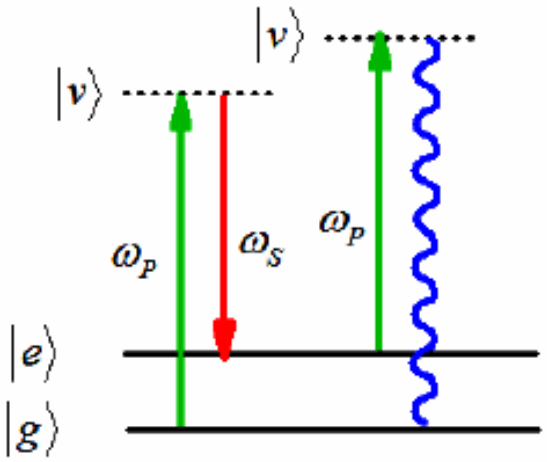
FIGURE 10.5.1 Various Raman scattering processes: (a) spontaneous Raman scattering; (b) stimulated Raman scattering; (c) coherent anti-Stokes Raman scattering (CARS)

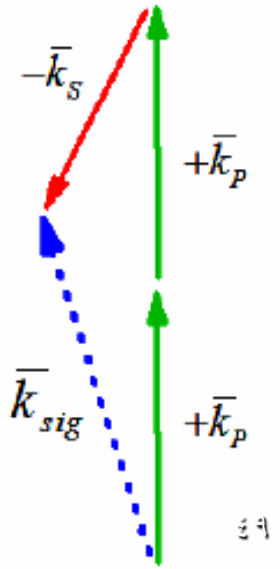
In either of these processes, two laser beams at frequencies ω_1 and $\omega_2 < \omega_1$ are applied to the Raman medium, and a beam at a new frequency is generated by the interaction. In the process of coherent anti-Stokes Raman scattering (CARS), illustrated in part (c) of the figure, an output is created at frequency $2\omega_1 - \omega_2$ as a consequence of the susceptibility $\chi_F^{(3)}(\omega_a = \omega_1 - \omega_2 + \omega_1)$. In the process of coherent Stokes Raman scattering (CSRS), illustrated in part (d) of the figure, an output is created at frequency $2\omega_2 - \omega_1$ as a consequence of the susceptibility $\chi_F^{(3)}(\omega_S = \omega_2 + \omega_2 - \omega_1)$.

CARS (Coherent Anti-Stokes Raman Scattering)

Used to drive ground state vibrations with optical pulses or cw fields.

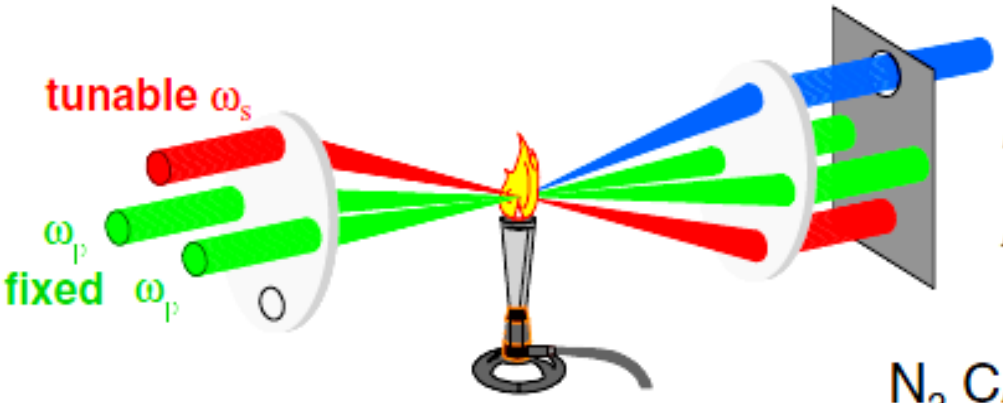
- Two fields, with a frequency difference equal to a vibrational transition energy, are used to excite the vibration.
- The first field is the "pump" and the second is the "Stokes" field.
- A second interaction with the pump frequency lead to a signal that radiates at the anti-Stokes frequency: $\omega_{sig} = 2\omega_P - \omega_S$ and the signal is observed background-free next to the transmitted pump field: $\bar{k}_{sig} = 2\bar{k}_P - \bar{k}_S$.





L

Coherent anti-Stokes Raman Scattering; CARS

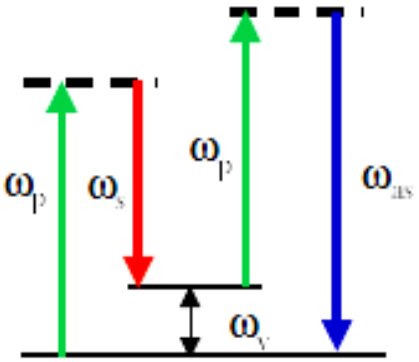


$\omega_{as} = 2\omega_p - \omega_s$

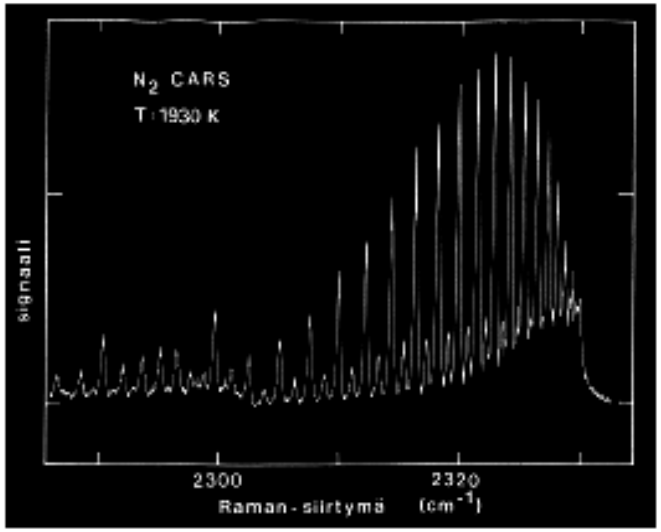
$\vec{k}_{as} = 2\vec{k}_p - \vec{k}_s$

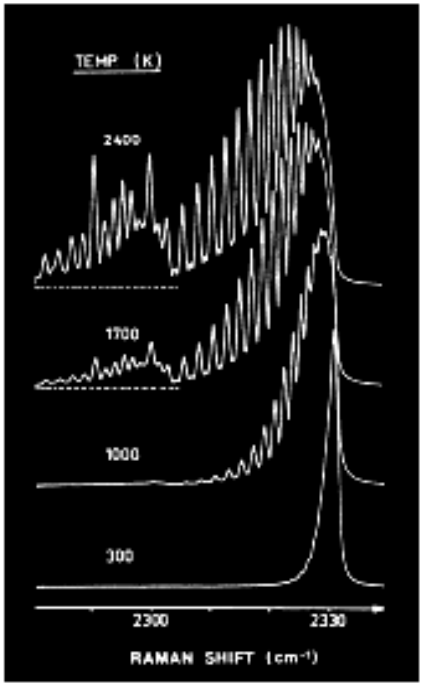
$I_a \propto N^2 I_p^2 I_s$

Combustion studies
- ✓ Temperature measurements
- ✓ High spatial and temporal resolution
- ✓ Efficient reduction of background signal caused by fluorescence



N_2 CARS spectra





The coherent scattering of these photons in CARS can be applied with very short pulses because there is almost no time delay from the electronic transitions and allows highly sensitive measurements if phase matching is achieved. In addition a high spatial resolution in the μm range is possible if the spatial overlap of the two pump beams is designed for this purpose. CARS is a four-wave mixing (FWM) process which is described in general in Sect. 5.9.2 (p. 335).

Phase matching is achieved if the momentum of the four attended photons are conserved and thus the wave vectors of the incident laser light $\boldsymbol{k}_{\mathrm{inc}}$ and of the Raman Stokes light $\boldsymbol{k}_{\mathrm{Stokes}}$ and the anti-Stokes light $\boldsymbol{k}$ have to fulfill the angle condition of Fig. 4.45.

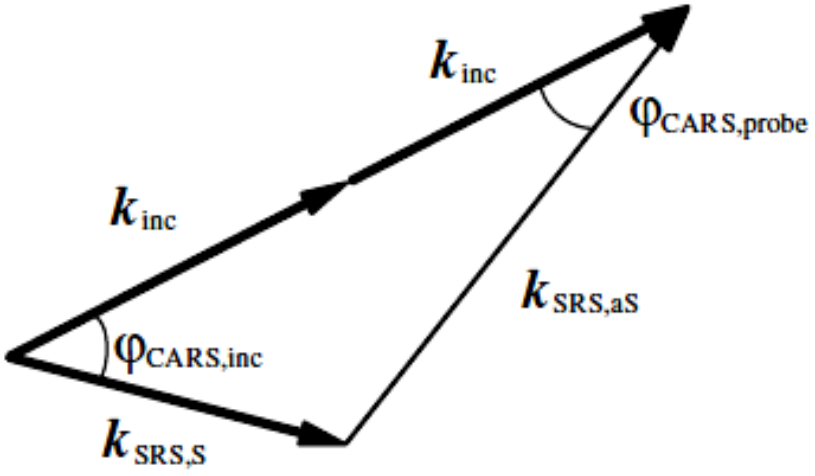


Fig. 4.45. Phase matching of the incident laser light and the generated Stokes and anti-Stokes Raman light in CARS experiments

Therefore the two incident laser beams have to be enclosed in the angle $\varphi_{\mathrm{CARS,inc}}$ and the anti-Stokes Raman light beam can be observed at the angle $\varphi_{\mathrm{CARS,probe}}$ to the laser beam propagation with $\boldsymbol{k}_{\mathrm{inc}}$ in the forward direction.

Thus CARS allows highly sensitive measurements of the anti-Stokes Raman signal in a spatial direction with no background light. The two strong pump lasers will populate the excited vibrational level and therefore highly efficient anti-Stokes Raman scattering will occur.

The scattering intensity $I_{\mathrm{CARS,aS}}$ is proportional to:

$$I_{\mathrm{CARS,aS}} \propto I_{\mathrm{inc}}^2 I_{\mathrm{SRS,S}} N_{\mathrm{mat}}^2 \tag{4.208}$$

with the pump laser intensities I_{inc} of the incident and $I_{\mathrm{SRS,S}}$ of the tuned light and particle density N_{mat}. Even continuously operating (cw) lasers can be used and then very high spectral resolution can be obtained because of the possible narrow band width of these lasers.

The scattering efficiency can be increased by many orders of magnitude if the pump laser photon energy matches the electronic transitions of the material (*resonant CARS*) [e.g. 4.623]. In this case the virtual Raman levels of the energy schemes above will be real energy states of the matter. Absorption will take place and thus the interaction length L and/or concentration N are limited by the maximum optical absorption of approximately $\sigma_{\mathrm{pump}} NL < 1$.

Optical Phase Conjugation (OPC)

(also called time reversal or wavefront reversal)

It is a nonlinear process used to remove the effects of aberrations from certain types of optical systems

It can exactly reverse the propagation direction and phase variation of a beam of light. The reversed beam is called a **conjugate** beam.

Then OPC is generation of a time-reversed replica of the wave, like a mirror, and

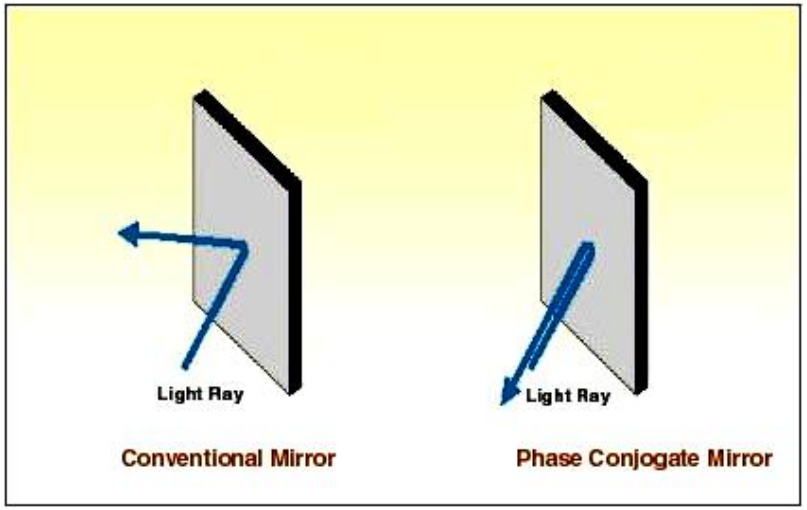
$$E = A \cos(\omega t - kz - \phi)$$

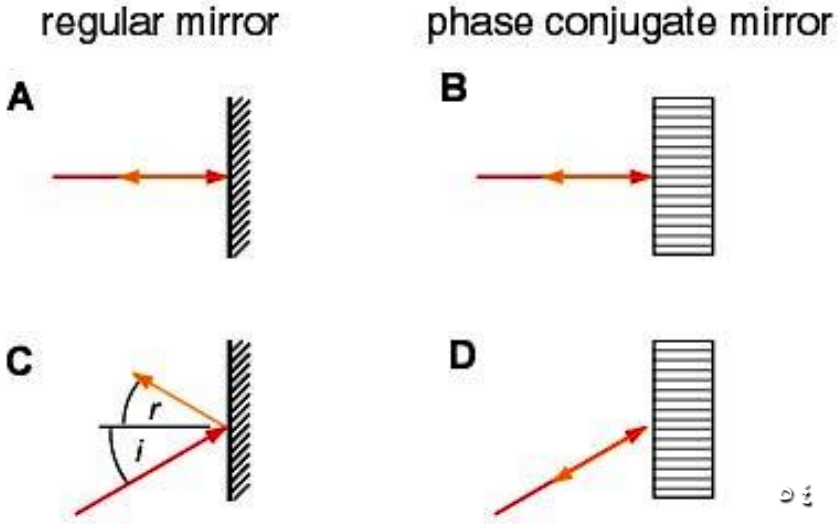
$$E_c = A \cos(\omega t + kz + \phi)$$

Phase Conjugate Mirror (PCM)

The PCM reflects incident light back towards where it came from, but it does so in a different way than a regular mirror.

In a regular mirror, light that strikes the mirror normal to its surface, is reflected straight back the way it came (A). This is also true of PCM (B). When the light strikes a normal mirror at an angle, it reflects back in opposite direction, such that the angle of incidence is equal to the angle of reflection (C). In PCM, light is always reflected straight back the way it came from, no matter what the angle of incidence (D).





Conventional mirror

Phase conjugating mirror

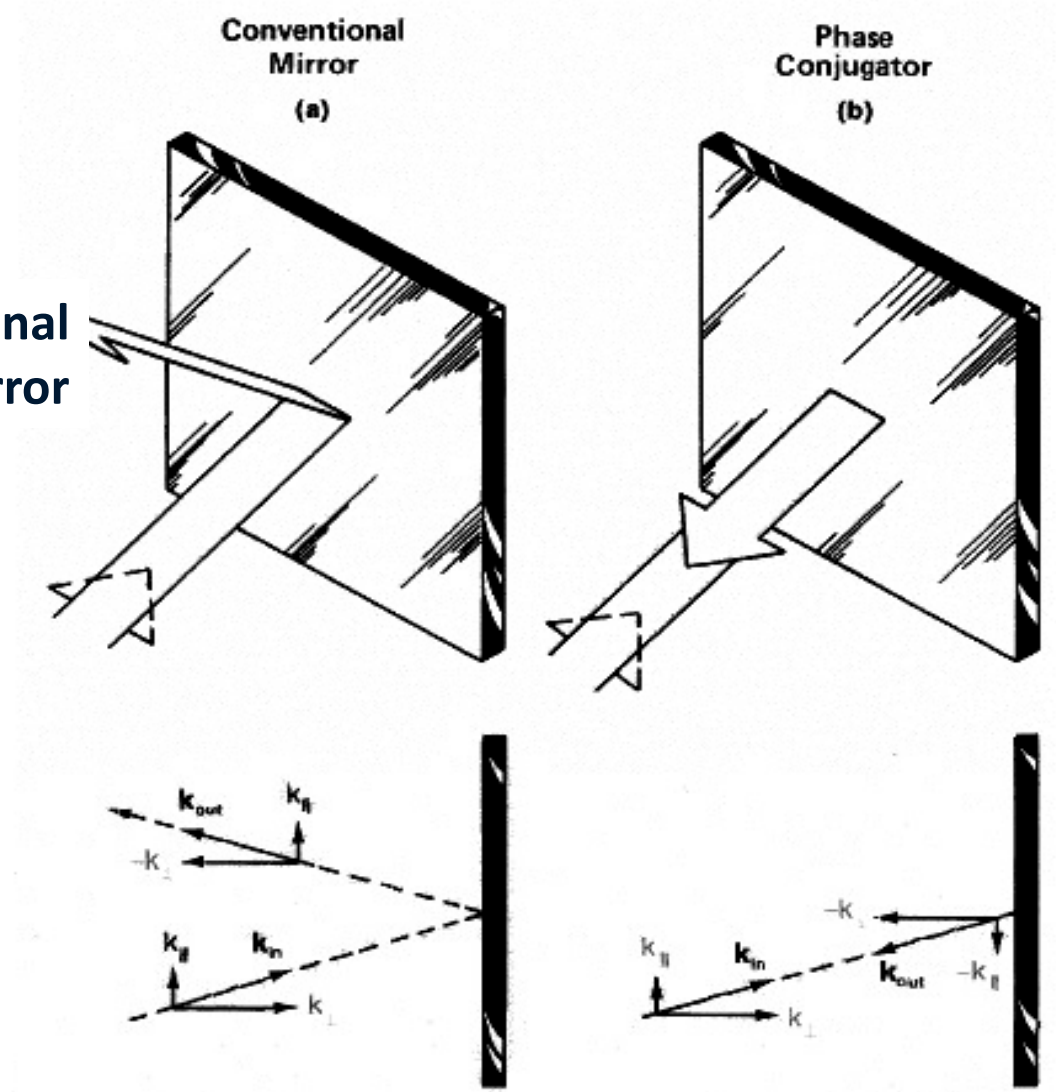
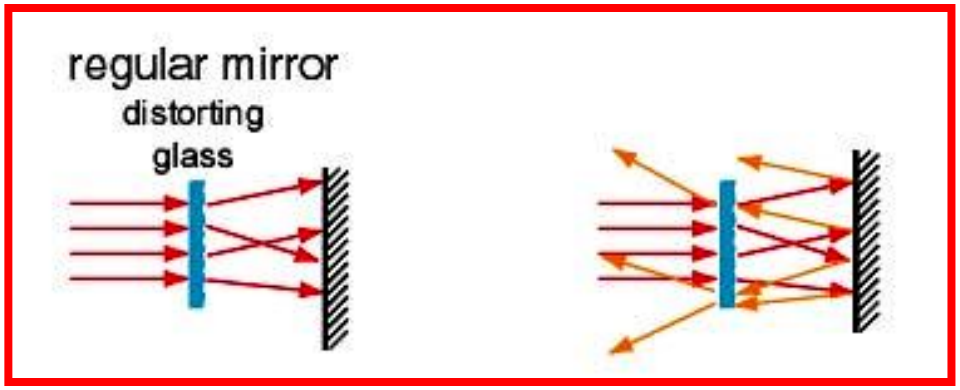
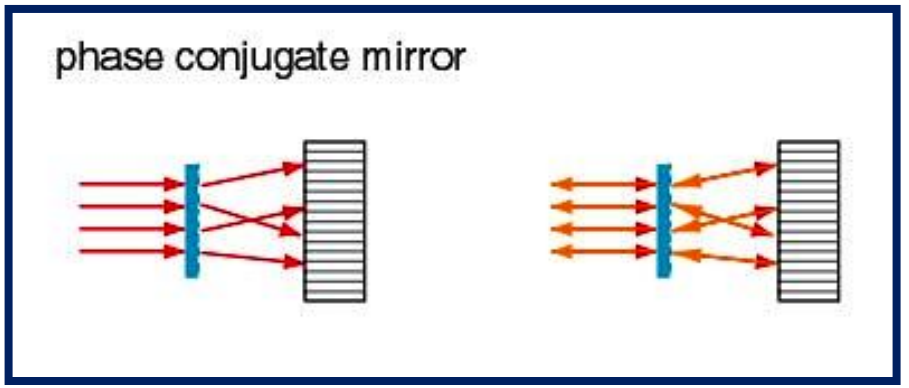


Fig. 1. (a) A conventional mirror reflects light by inverting only the normal component $\mathbf{k}_{\perp}$ *of the beam's propagation vector* **k**. *This process leads to the law that the angle of incidence equals the angle of reflection and allows the direction of the reflected beam to be altered by changing the tilt of the mirror. (b) A phase conjugator reflects light by inverting all components so that the propagation vector changes sign* ($\mathbf{k}_{out} = -\mathbf{k}_{in}$). *In this case, regardless of the tilt of the mirror, the reflected light exactly retraces the path of the incoming beam.*

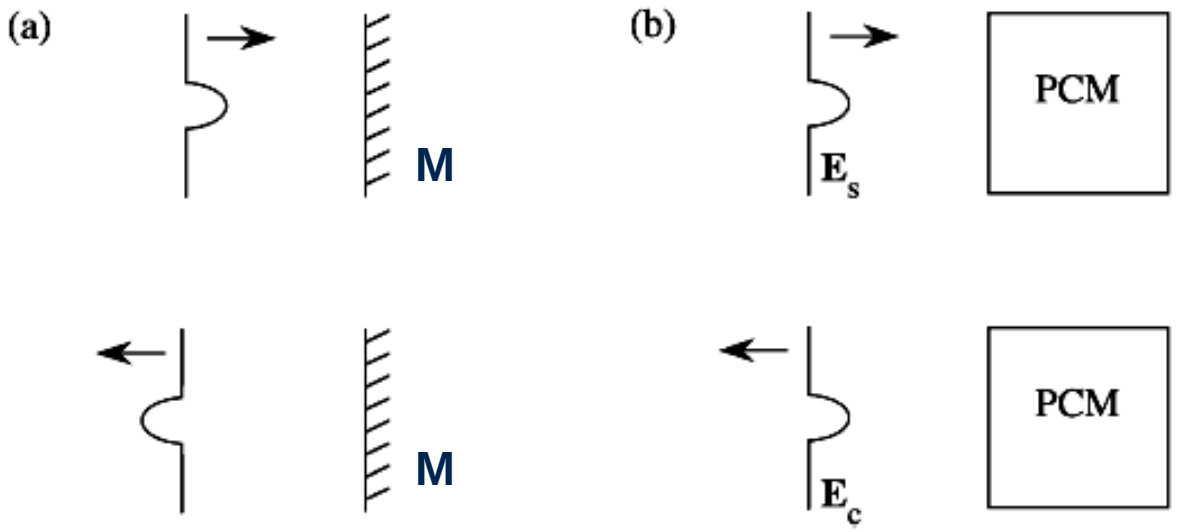




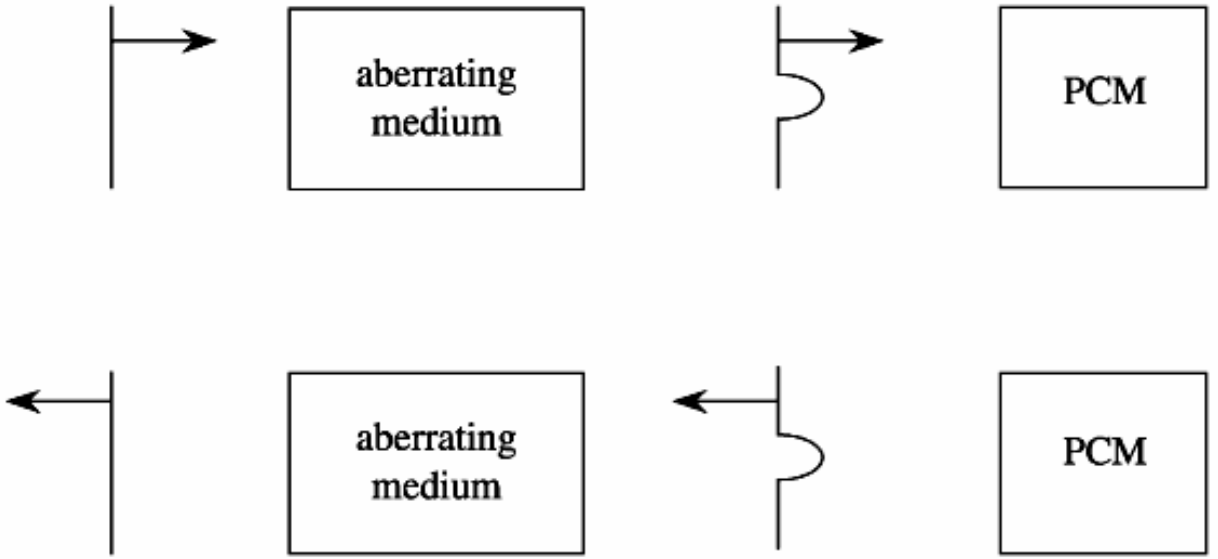
This difference has significant consequences, e.g., if we place an irregular distorting glass in the path of a beam of light, the parallel rays get bent in random directions, and after reflection from a normal mirror, each ray of light is bent even farther, and the beam is scattered.

With a PCM, each ray is reflected back in the direction it came from. This reflected conjugate wave therefore, propagates backwards through the distorting medium, and essentially "un-does" the distortion, and returns to a coherent beam of parallel rays travelling in the opposite direction.

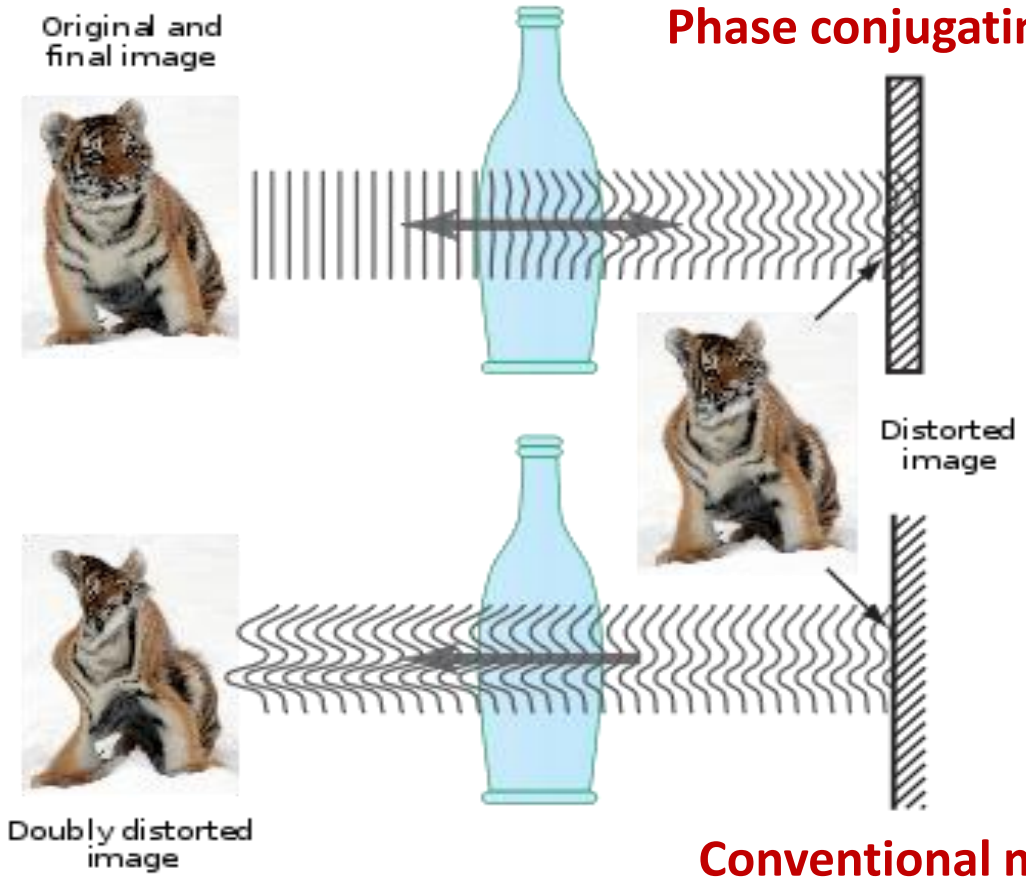
Phase Conjugation



Reflection from (a) an ordinary mirror and (b) a phase-conjugate mir-



Aberration correction by optical phase conjugation.



Comparison of a phase conjugate mirror with a conventional mirror. With the phase conjugate mirror the image is not deformed when passing through an aberrating element twice.

4.5.14 Optical Phase Conjugation via Stimulated Scattering

The conjugation of the phase of an optical wave is equivalent to inversion of the wave front of the light beam [4.626–4.629]. It allows the realization of phase conjugating mirrors (PCM's) which are, for example, used in lasers for improving the beam quality, as described in Sects. 6.6.12 (p. 416) and 6.11.3 (p. 480) [4.630, 4.631]. Further examples can be found in [4.632–4.692]. If the incident light beam is described by the electric field vector E_{inc}:

$$\text{incident light} \quad \boldsymbol{E}_{\mathrm{inc}}(\boldsymbol{r},t) = \mathrm{Re}\{\boldsymbol{E}_0(\boldsymbol{r})\,\mathrm{e}^{\mathrm{i}2\pi\nu t}\} \tag{4.210}$$

with the complex amplitude

$$\boldsymbol{E}_0(\boldsymbol{r}) = \boldsymbol{A}_0(\boldsymbol{r})\,\mathrm{e}^{-\mathrm{i}(k\boldsymbol{r}+\varphi)} \tag{4.211}$$

the phase conjugate is given by:

$$\text{phase conjugate} \quad \boldsymbol{E}_{\mathrm{phconj}}(\boldsymbol{r},t) = \mathrm{Re}\{\boldsymbol{E}_0^*(\boldsymbol{r})\,\mathrm{e}^{\mathrm{i}2\pi\nu t}\} \tag{4.212}$$

with the complex conjugated amplitude:

$$\text{conjugated amplitude} \quad \boldsymbol{E}_0^*(\boldsymbol{r}) = \boldsymbol{A}_0(\boldsymbol{r})\,\mathrm{e}^{+\mathrm{i}(k\boldsymbol{r}+\varphi)} \tag{4.213}$$

where the sign of the spatial phase is changed. The conjugate can also be written as:

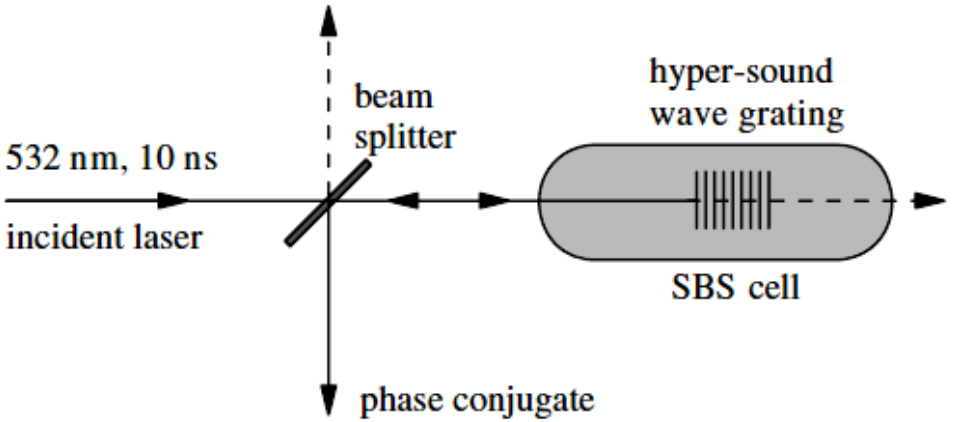
$$\boldsymbol{E}_{\mathrm{phconj}}(\boldsymbol{r},t) = \mathrm{Re}\{\boldsymbol{E}_0(\boldsymbol{r})\,\mathrm{e}^{-\mathrm{i}2\pi\nu t}\} \tag{4.214}$$

with the unchanged spatial amplitude:

$$\boldsymbol{E}_0(\boldsymbol{r}) = \boldsymbol{A}_0(\boldsymbol{r})\,\mathrm{e}^{-\mathrm{i}(\boldsymbol{k}\boldsymbol{r}+\varphi)} \tag{4.215}$$

but there is a change of sign of the temporal phase. This corresponds formally to a change in time direction indicating that the phase front is moving perfectly backwards. It does not indicate a time direction change for the pulse shape or in general!

This phase conjugation can be achieved with nonlinear back-reflection of the beam via stimulated scattering, e.g. via stimulated Brillouin scattering (SBS) as described in Sect. 5.9.2 (p. 335) or via four-wave mixing (FWM) (see e.g. [4.632–4.642]). Such a volume reflector is called a phase conjugating mirror (PCM). Stimulated Brillouin scattering such as a self-pumped process allows very easy realization of this process as can be seen in Fig. 4.47 (p. 251).



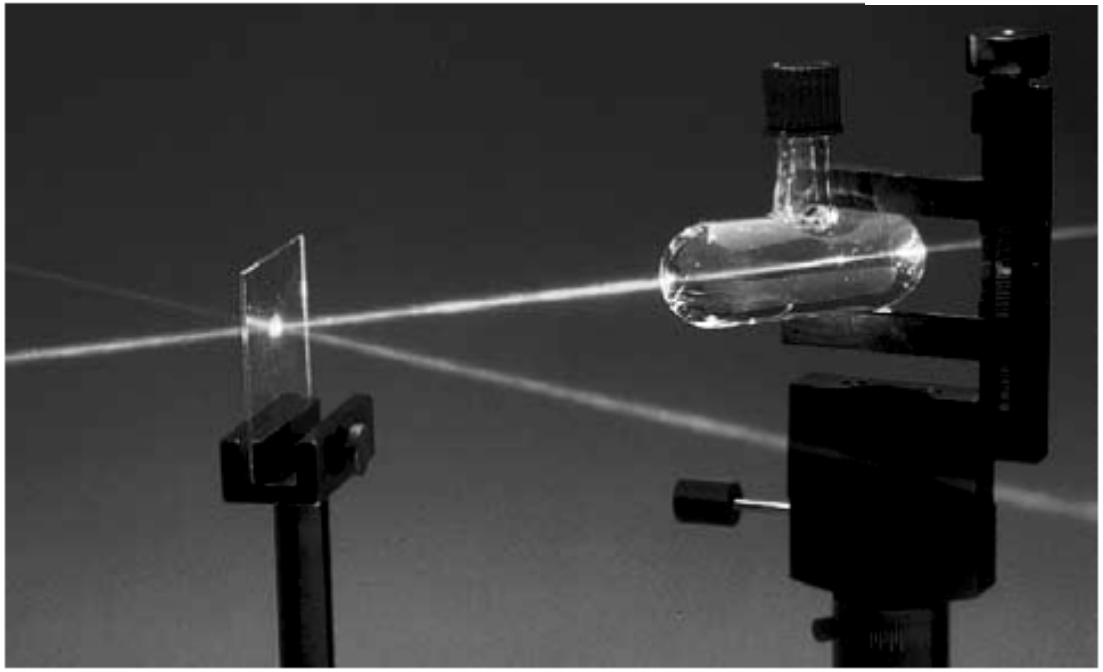


Fig. 4.47. Optical phase conjugation of a frequency-doubled Nd:YAG laser beam with a pulse duration of 20 ns and a pulse energy of 20 mJ via SBS in a cell filled with acetone. The beam was photographed in smoky air. The incident light propagated from the left into the cell and is focused by the curved cell surface. The phase conjugated light is reflected towards the observer by the beam splitter (Foto:

The second harmonic light of a pulsed Nd:YAG laser beam which was used for being visible with a duration of 20 ns and a pulse energy of 20 mJ was focused by the curved entrance window of the glass cell into the liquid acetone. As can be seen in the picture, this material is transparent for low light powers at this wavelength in the green spectral region. In the focus the sound wave grating of the SBS is established and reflects in this simple demonstration experiment about 50% of the incident light as a PCM. The reflected light observable behind the beam splitter shows the same properties as the incident light beam although the imperfect cell window introduces severe phase distortions.

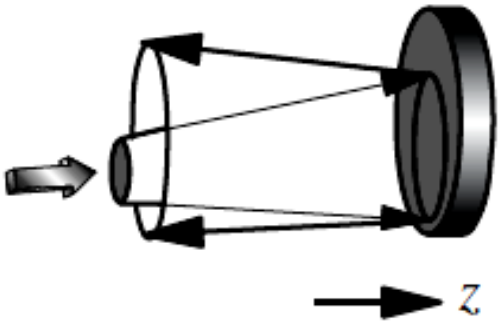
Such a phase conjugating mirror shows unusual properties compared to a conventional mirror (see Fig. 4.49, p. 252) with important applications in photonics especially in laser technology.

Most obviously the light is perfectly back-reflected by the PCM independent of the direction of the incident light beam. Thus not only is the z component of the wave vector inverted but so are all components in the PCM independent of their directions.

Conventional mirror

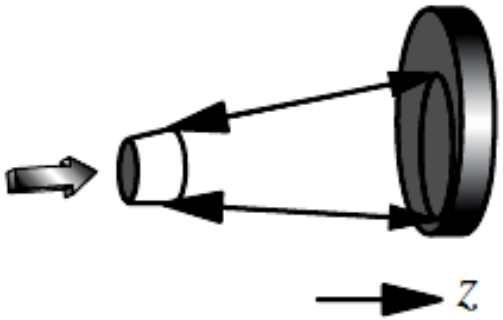
wave vector k after reflection

Phase conjugating mirror



$k_x^{out} = k_x^{in}$ $k_y^{out} = k_y^{in}$ $k_z^{out} = -k_z^{in}$

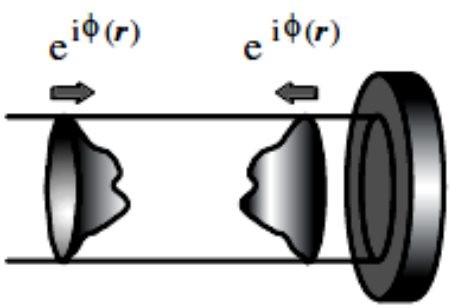
partial reverse wave vector $\boldsymbol{k}$



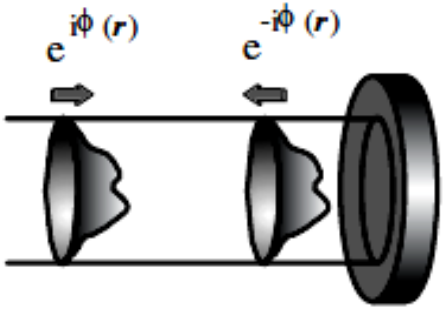
$k_x^{out} = -k_x^{in}$ $k_y^{out} = -k_y^{in}$ $k_z^{out} = -k_z^{in}$

complete reverse wave vektor $\boldsymbol{k}$

wave front after reflection



wavefront: $\phi_{out}(\boldsymbol{r}) = \phi_{in}(\boldsymbol{r})$



wavefront: $\phi_{out}(\boldsymbol{r}) = -\phi_{in}(\boldsymbol{r})$

Fig. 4.49. Reflectivity properties of a phase conjugating mirror (PCM) in comparison to conventional mirrors

A third important property of the PCM is the treatment of the polarization of the light (see Fig. 4.51, p. 253).

In PCMs the linear polarization of light is unchanged and for many PCM processes linear polarized light is most efficient. In complete phase conjugating mirrors the circular or elliptic polarization is conserved. Thus this type is called *vector phase conjugation*. It can be achieved in four-wave mixing (FWM) schemes, only. Phase conjugating mirrors based on stimulated scattering, e.g. SBS, will not conserve the spin direction of circular or elliptic polarization because the stimulation of the sound wave is an intensity effect and therefore does not contain all effects of the electric field vectors. Thus PCMs based on SBS will treat light polarization in the same way as conventional mirrors. As a consequence double pass arrangements with SBS-PCM can be very easily build with a polarizer and a Faraday

The incident and the phase conjugated beams can thus be separated by their different polarizations whereas their beam shape is almost exactly the same.

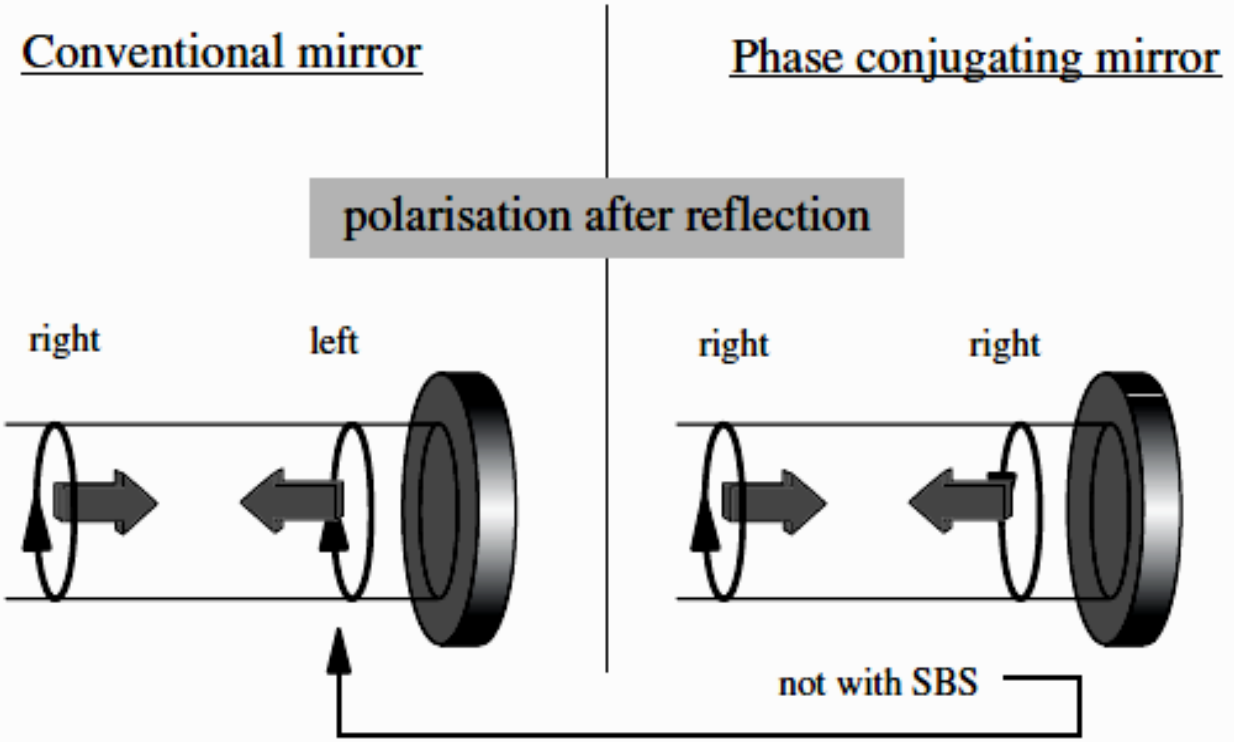


Fig. 4.51. Polarization of a light beam after reflection in a phase conjugating mirror (PCM) in comparison to a conventional mirror. With SBS the polarization is treated conventionally but with vector phase conjugation the polarization is conserved