

NLO Lecture 13: Raman und Brillouin Scattering

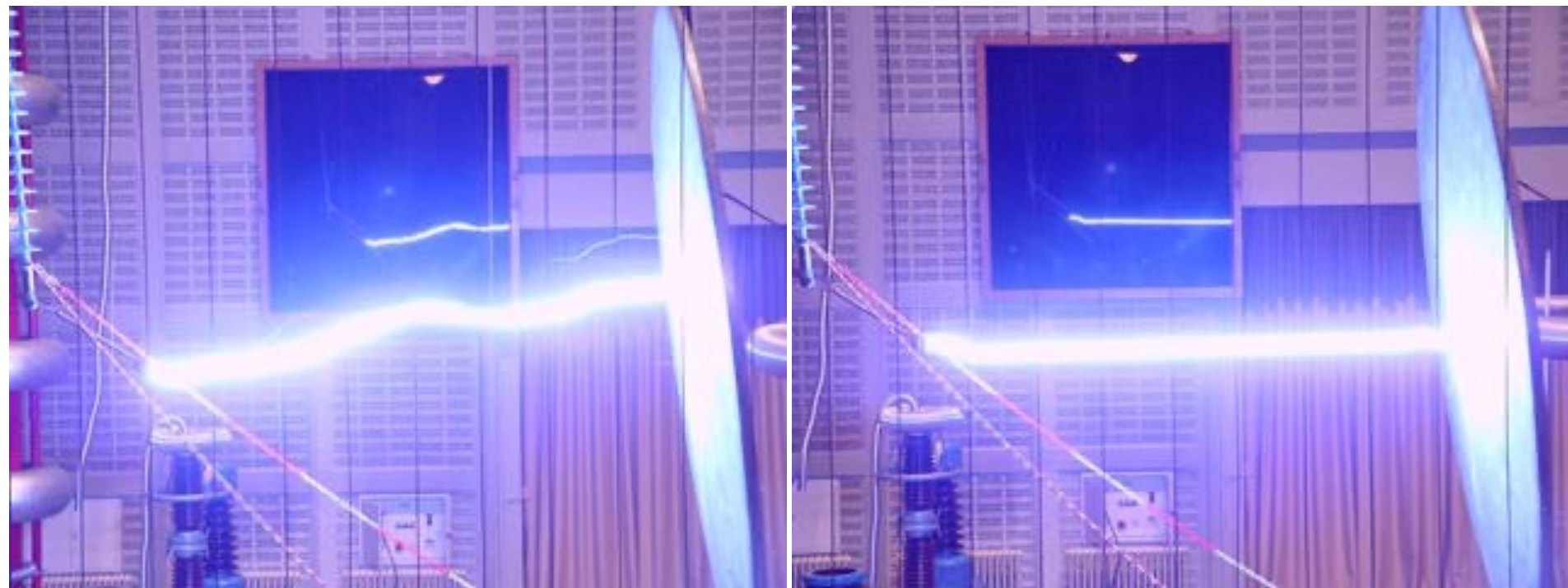
- 7 Third-order nonlinear effects (continued)**
- 7.6 Raman and Brillouin scattering**
- 7.6.1 Focusing**
- 7.6.2 Strong conversion**
- 7.6.3 Stimulated Brillouin scattering (SBS)**



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>] 3



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

7.6 Raman and Brillouin scattering

Stimulated Raman and Brillouin scattering is an important technique to investigate **low energy excitations in molecules and solids**

They permit **identification and study of those excitations**, **without direct coupling to the optical radiation**.

stimulated Raman scattering occurs 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 and Raman lasers.

Physical effect of Raman scattering:

Light propagating through a sample with polarization fluctuations can be **scattered in arbitrary direction and shifted in frequency**

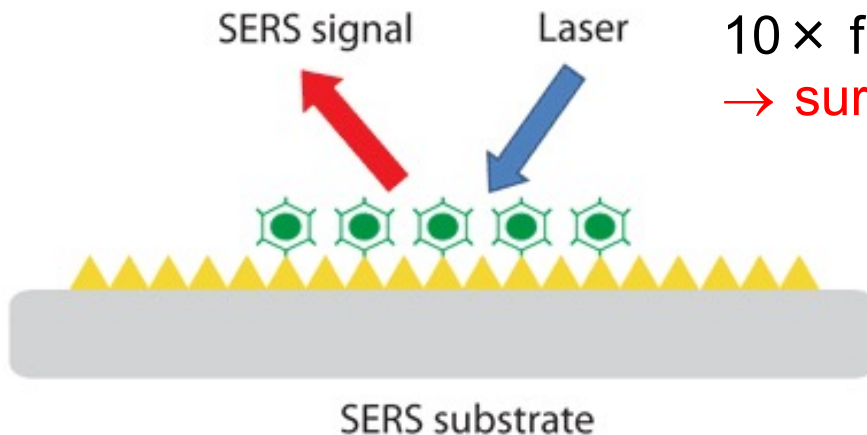
If the polarization fluctuations originate from **oscillations of a molecules** or **optical or acoustic phonons in a solid**, the process takes place via **absorption or emission of a phonon**, leading to an **Anti-Stokes or Stokes shift** of the scattered photon

absorption or emission of a phonon, leading to an Anti-Stokes or Stokes shift of the photon

$$\omega_{AS} = \omega_L + \Omega \quad (7.66)$$

$$\omega_S = \omega_L - \Omega, \quad (7.67)$$

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.



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

$$\alpha = \alpha_0 + \frac{\partial \alpha}{\partial Q} \cdot Q \quad (7.68)$$

a contribution to the nonlinear polarization of the form

$$P_{NL} = N \varepsilon_0 \frac{\partial \alpha}{\partial Q} Q E, \quad \text{we need } Q \quad (7.69)$$

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

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

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

$$F = -\frac{1}{2} \varepsilon_0 E^2 \frac{\partial \alpha}{\partial Q}. \quad \propto E^2 \quad (7.71)$$

The oscillation amplitude then satisfies the equation

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

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

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

With this we obtain for the stationary oscillation

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

with

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

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

$$\tilde{Q}(z, t) = \frac{\frac{\epsilon_0}{4m\Omega_0} \frac{\partial\alpha}{\partial Q} E_L E_S^*}{\Omega_0 - (\omega_L - \omega_S) + j\frac{\Gamma}{2}} e^{j\{(\omega_L - \omega_S)t - (k_L - k_S)z\}}. \quad (7.76)$$

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

$$\frac{\partial E_S}{\partial z} = -\frac{j\omega_s}{cn_s} N \frac{\partial\alpha}{\partial Q} \tilde{Q}^* E_L \quad \text{all other beat terms ignored here} \quad (7.77)$$

or

$$\frac{\partial E_S}{\partial z} = \frac{j\omega_s \epsilon_0 N \left| \frac{\partial\alpha}{\partial Q} \right|^2 |E_L|^2}{4\Omega_0 m c n_s \left\{ (\omega_L - \omega_S) - \Omega_0 + \frac{j\Gamma}{2} \right\}} E_S. \quad (7.78)$$

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

$$I_S = \frac{n_s}{2} \sqrt{\frac{\epsilon_0}{\mu_0}} |E_S|^2 \quad (7.79)$$

follows

$$I_S(\ell) = I_{S0} \exp\{g I_L \ell\} \quad (7.80)$$

with the Raman gain

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



C. V. Raman
(Nobel Prize
1930)

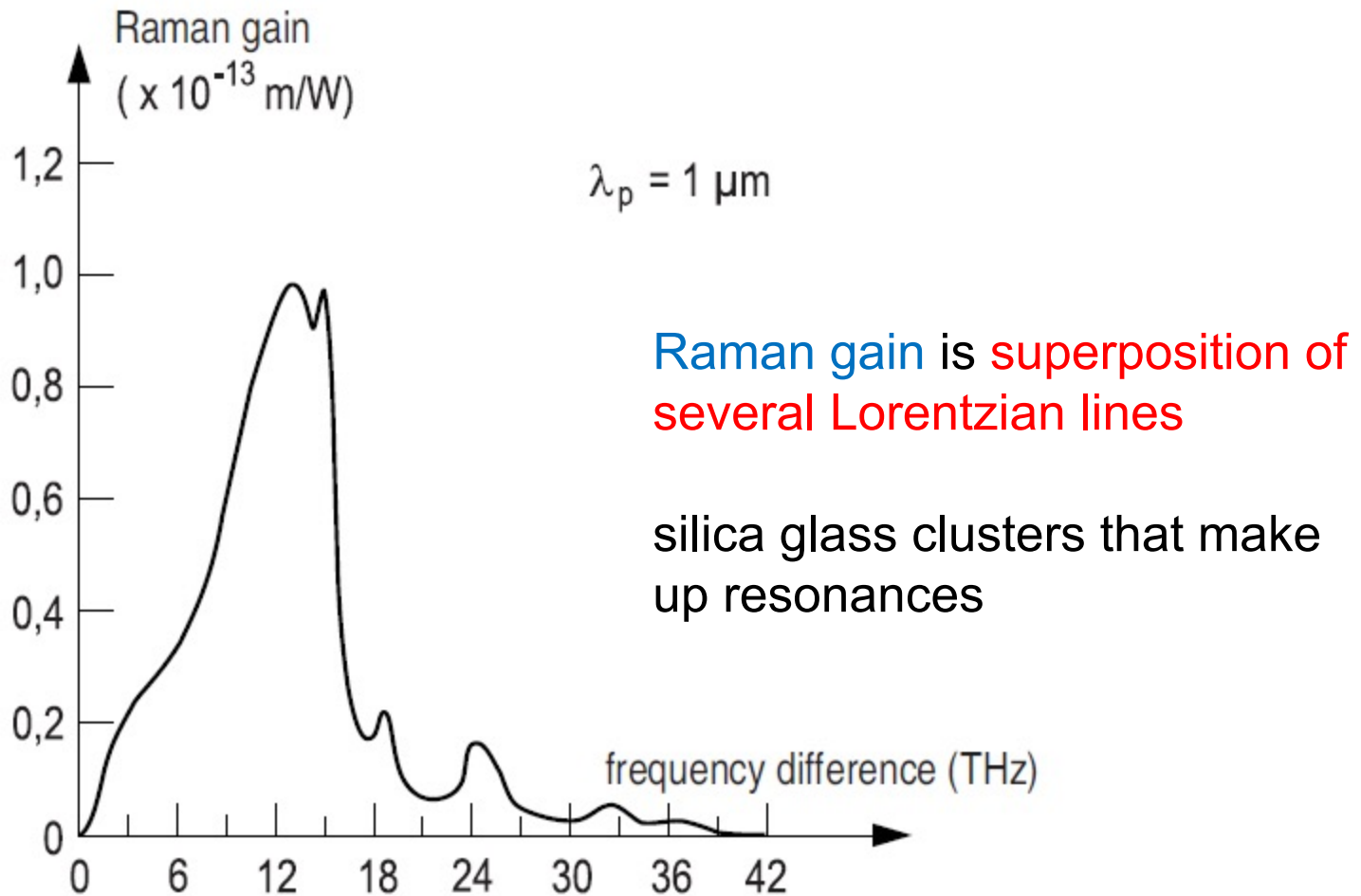


Figure 7.6: Measured Raman amplification gain of melted quartz at a pump wavelength of $1 \mu\text{m}$. The horizontal axis shows difference frequency between laser and Stokes line.

7.6.1 Focusing

For the total gain, we obtain

$$G = \int_0^\ell g I_L dz.$$

For a Gaussian beam, we then obtain, independent of focusing, the on-axis gain (with intensity $I_{L,\max}(z) = \frac{P_L}{\pi w_0^2 (1 + \frac{z^2}{b^2})}$ and $b = \pi w_0^2 / \lambda$)

$$G_b = \int_{-\infty}^{\infty} g I_{L,\max} dz = \frac{\pi b}{\pi w_0^2} g P_L = \frac{\pi}{\lambda} g P_L.$$

We again find that the effective Raman gain, which results from a $\chi^{(3)}$ -effect, in a volume is independent of focusing, as the effective interaction length is proportional to the confocal parameter b .

In contrast, we obtain for the Raman gain in a glass fiber with core radius r_0 and length ℓ

$$G_f = \frac{g P_L}{\pi r_0^2} \ell.$$

Thus the waveguiding structure enhances the Raman gain by a factor

$$\frac{G_f}{G_b} = \frac{\ell \lambda}{\pi^2 r_0^2}.$$

For a given situation, this ratio can easily amount to 10^6 , e.g., for $\ell = 40$ m, $r_0 = 2 \mu\text{m}$ and $\lambda = 1 \mu\text{m}$. In the real world, the effective fiber length is limited by the absorption length $1/\alpha$ for the pump light. This corresponds to several kilometers, $P_L = P_{L0} \exp[-\alpha z]$. For a long fiber ($L \gg 1/\alpha$), we obtain

$$G_f = \frac{P_{L0}}{\pi\tau_0^2} (g/\alpha).$$

For large gain, we can neglect the losses at the Stokes frequency. The light at the Stokes frequency emerges from initial noise, this is referred to as spontaneous scattering. A quantum treatment reveals that the power at the amplifier output originating from spontaneous processes is equivalent to 1 photon per mode at the input of the amplifier.

The output power at the Stokes frequency originating from input noise is becoming comparable to the pump power at a gain of [8]

$$G_f \cong 16,$$

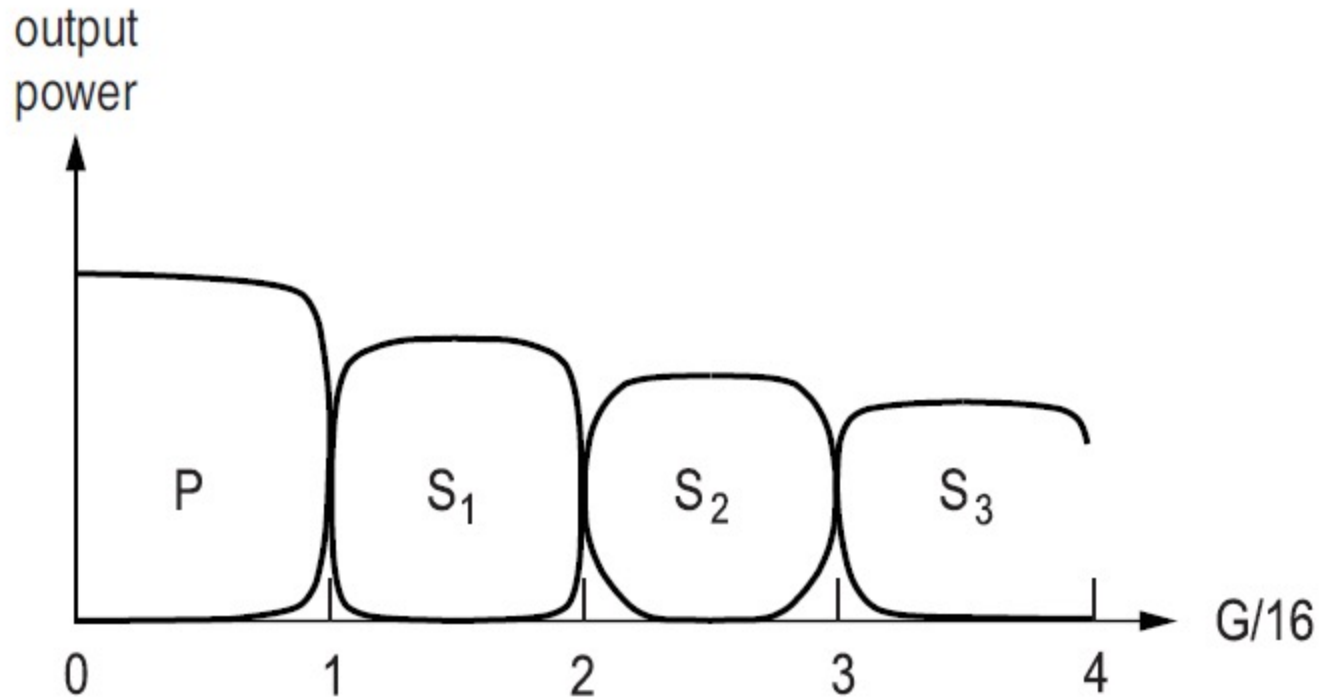
and the required pump power is then given by

$$P_{L0} \cong 16 (A_{eff}) (\alpha/g).$$

For a maximum Raman gain of $g = 0.1/\text{m} \cdot (\mu\text{m}^2/A_{eff}) \cdot (P/W)$ from Fig. 7.6 and 0.2 dB damping, corresponding to an absorption length of 20 km, at the communications wavelength $1.55 \mu\text{m}$, and $A_{eff} = 50 \mu\text{m}^2$, we obtain a threshold power of $P_{L0} = 600$ mW.

7.6.2 Strong conversion

For input powers far above threshold, it is possible to create several Stokes lines as shown in Fig. 7.7. If the total power of the pump pulse (P) is transferred to the first Stokes line (S_1), then in the next fiber section a conversion to (S_2) occurs, and so on.



7.6.3 Stimulated Brillouin scattering

Brillouin scattering: scattering of light on acoustic waves

Again as for Raman scattering, fluctuations of polarizations, but now caused by acoustic waves, give rise to spontaneous and stimulated scattering.

First observation of Stimulated Brillouin scattering (SBS):

R. Y. Chiao, C. H. Townes, and B. P. Stoicheff, Phys. Rev. Lett. 12, 592 (1964).

For strong pump fields: very efficient frequency conversion

In contrast

to Raman-active molecule oscillations or optical phonons, acoustic waves or phonons propagate with a velocity v_a , thus the wave number κ and frequency Ω of the acoustic wave are related by

$$\kappa = \frac{\Omega}{v_a}. \quad (7.82)$$

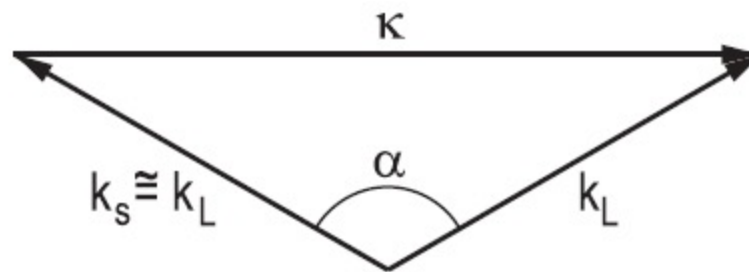
In the scattering process, momentum and energy must be conserved

$$\mathbf{k}_S = \mathbf{k}_L - \boldsymbol{\kappa}. \quad (7.83)$$

$$\omega_S = \omega_L - \Omega. \quad (7.84)$$

From the second condition follows

$$k_S = \frac{n_S}{n_L} k_L - \frac{n_S v_a}{c} \kappa. \quad (7.85)$$



Since acoustic frequencies (kHz - 100 GHz) are much smaller than optical frequencies (300 THz), it follows $\omega_S \approx \omega_L$ and thus $k_S \approx \frac{n_S}{n_L} k_L \approx k_L$. The scattering geometry for SBS thus looks as depicted in Fig. 7.8. Furthermore, the sound velocity is much smaller than that of light, $v_a \ll c$, such that we approximately have $v_a/c \approx 10^{-5}$. The wavelength of the sound wave can thus easily become of the same order of magnitude as that of light, therefore in general arbitrary angles α are possible, i.e.,

$$\kappa \cong 2k_L \sin \frac{\alpha}{2} \quad \Rightarrow \quad \sin \frac{\alpha}{2} \cong \frac{\kappa}{2k_L} = \frac{\lambda_L}{2\Lambda n}, \quad (7.86)$$

where λ_L is the wavelength of light in the medium with refractive index n and Λ the wavelength of the sound wave. This is the same condition as the Bragg condition for scattering of X-rays from crystals. Again the longest interaction length can be achieved in a guided collinear geometry of laser and Stokes because from Eq. (7.86) and $\alpha = 0$, it follows that $\Omega_B = \kappa = 0$. However, it is possible in backscattering geometry, $\alpha = \pi$, it then holds

$$\Lambda = \frac{\lambda_L}{2n}. \quad (7.87)$$

Thus the wavelength of light in combination with the phase velocity of the sound wave and the refractive index of the material determine the frequency of the interacting sound wave via

$$\Omega_B = \frac{4\pi n v_a}{\lambda_L}. \quad (7.88)$$

For infrared light in glass with $\lambda_L = 1.55 \mu\text{m}$, $n = 1.5$, and $v_a = 5.96 \text{ km/s}$, we obtain an SBS frequency of $f_B = \Omega_B/2\pi = 11.5 \text{ GHz}$.

We now want to consider again the acousto-optical coupling. The amplitude of elongation is $Q(t, z) = Qe^{j(\Omega t - \kappa z)}$, which satisfies

$$\frac{\partial^2 Q(z, t)}{\partial t^2} + \Gamma \frac{\partial Q(t, z)}{\partial t} - v_a^2 \frac{\partial^2 Q(t, z)}{\partial z^2} = \frac{1}{\rho} F(t, z). \quad (7.89)$$

where ρ is the density of the medium and Γ a damping constant. The light field now couples to a wave and not to localized oscillations. In analogy to the Raman effect, the driving force is given by

$$F(t, z) = \frac{1}{2} \varepsilon_0 E^2 \frac{\partial \alpha}{\partial Q}. \quad (7.90)$$

However, we point out that here the polarizability is modulated by the stress created in the medium which is only one consequence of the oscillation elongation Q . In general, a change in the refractive index ellipsoid resulting from stress is described by the elasto-optical coefficient p

$$\Delta(1/n^2) = pS \quad \Rightarrow \quad \Delta n = -\frac{n^3}{2} pS, \quad (7.91)$$

or

$$\Delta \alpha = \Delta(\varepsilon/\varepsilon_0) = 2n\Delta n = -n^4 pS. \quad (7.92)$$

The stress results from the elongation Q according to

$$S = \frac{\partial Q}{\partial z} = -j\kappa Q, \quad (7.93)$$

such that

$$\frac{\partial \alpha}{\partial Q} = j\kappa n^4 p. \quad (7.94)$$

For slowly varying amplitudes (SVEA) and $\Omega^2 = v_a^2 \kappa^2$, Eq. (7.89) for the oscillation amplitude simplifies to

$$\frac{\partial Q}{\partial t} + \frac{\Gamma}{2}Q + v_a \frac{\partial Q}{\partial z} = \frac{\varepsilon_0 \kappa n_s^4 p}{4\Omega \rho} E_L E_S^*. \quad (7.95)$$

For the case of strong damping, e.g., in glass, a 50-GHz phonon only propagates only 12 μm , in the stationary state, $\partial/\partial t = \partial/\partial z = 0$, it follows

$$Q = \frac{\varepsilon_0 \kappa n_s^4 p}{2\Omega \rho \Gamma} E_L E_S^*. \quad (7.96)$$

The nonlinear polarization then is

$$P_{NL}(\omega_s) = \varepsilon_0 \frac{\partial \alpha}{\partial Q} Q^* \cdot E_L = j\kappa \varepsilon_0 n_s^4 p Q^* E_L. \quad (7.97)$$

The backwards scattered Brillouin radiation then grows exponentially according to

$$\frac{\partial E_S}{\partial z} = \frac{j\omega_s}{2cn_s\varepsilon_0} P_{NL} = -\frac{\kappa^2 n_s^7 p^2 \omega_s \varepsilon_0}{4c_0 \Omega \rho \Gamma} |E_L|^2 E_S$$

or

$$\frac{\partial I_S}{\partial z} = -g I_L I_S$$

with

$$g = \frac{8\pi^2 n_s^6 p^2}{\lambda_s^2 c_0 \rho v_a \Gamma} = \frac{4\pi n_s^6 p^2}{\lambda_s^2 c_0 \rho v_a \Delta\nu_B},$$

where $\Delta\nu_B = \Gamma/2\pi$ is the FWHM bandwidth for Brillouin scattering. Due to the narrower bandwidth $\Delta\nu_B$ on the order of a few to 100 MHz, Brillouin amplification is typically much stronger than Raman amplification. However, only pump light within a bandwidth $\Delta\nu_B$ contributes to Brillouin amplification at a certain frequency. Therefore, by using pulses with durations below 1 ns, whose spectrum is narrower than 300 MHz, one can suppress Brillouin scattering. For the propagation of ps (or even shorter) pulses, Brillouin scattering plays often no role. In contrast, when using narrowband lasers with linewidths below 10 MHz, the threshold for strong Brillouin scattering is reduced to a few mW. The threshold for Brillouin backscattering is ~ 20 .