Since the advent of laser spectroscopy a large part of the research effort in Rayleigh and Brillouin scattering (RB scattering) has been devoted to the study of excitations in fluids (for discussions, see Mountain 1966a, Fabelinskii 1968, Benedek 1968, McIntyre and Sengers 1968, Fleury and Boon 1973, Berne and Pecora 1976). Some of the concepts developed in the study of fluids have relevance to solids and will be briefly outlined here. Because the sound waves studied in light scattering are of long wavelength, the microscopic structure of fluids can be ignored to a first approximation. A thermodynamic treatment of fluctuations in a hydrodynamic medium, regarded as a continuous isotropic dielectric, leads to a spectrum of scattered light consisting of two types (Figure 8.1):

1. A quasielastic Rayleigh component centered at ω_I due to nonpropagating entropy fluctuations.
2. A Brillouin doublet symmetrically located about the unshifted line and separated from it by a frequency equal to that of a compressional sound wave propagating through the fluid.

The fluctuations in the susceptibility that give rise to RB scattering are due to variations in thermodynamic quantities, such as density and temperature. The state of a fluid in thermodynamic equilibrium consisting of a single constituent can be described by two variables, for example, the pressure $P(\mathbf{r},t)$ and the entropy $S(\mathbf{r},t)$ so that the effect of fluctuations in these variables on the susceptibility can be expressed as

$$\delta\chi(\mathbf{r},t)=\left(\frac{\partial\chi}{\partial P}\right)_S \delta P(\mathbf{r},t)+\left(\frac{\partial\chi}{\partial S}\right)_P \delta S(\mathbf{r},t). \tag{8.1}$$

Using (1.71) we find that the differential cross section for scattering of light by a fluid is

$$\frac{d^2\sigma}{d\Omega\, d\omega_S}=\frac{\omega_I^4 \upsilon V}{16\pi^2 c^4}\langle|\delta\chi|^2\rangle_\omega \cos^2\phi, \tag{8.2}$$

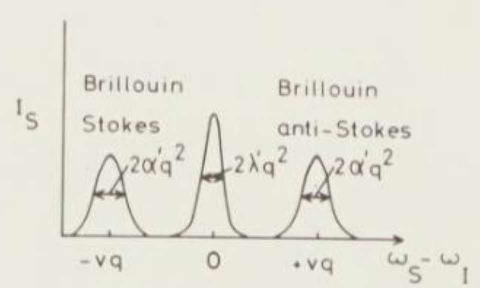

Figure 8.1 Schematic representation of Rayleigh and Brillouin spectra.

where ϕ is the scattering angle. Evaluation of $\langle|\delta\chi|^2\rangle_\omega$ leads to the result (e.g., see Cummins and Gammon 1966)

$$\frac{d\sigma}{d\Omega}=\frac{\omega_I^4 \mathfrak{v} k_B T}{16\pi^2 c^4}\left[\beta_S\rho^2\left(\frac{\partial\chi}{\partial\rho}\right)_S^2+\frac{T}{\rho C_P}\left(\frac{\partial\chi}{\partial T}\right)_P^2\right]\cos^2\phi, \tag{8.3}$$

where β_S is the adiabatic compressibility, ρ is the density, and C_P is the specific heat per unit mass. A formula of the type (8.3) was first obtained by Einstein (1910).

To a good approximation sound waves are pressure (density) fluctuations at constant entropy and the first term in the brackets in (8.3) is associated with the excitation of the Brillouin doublet corresponding to two sound waves with the same frequency travelling in opposite directions (Brillouin 1914, 1922). The second term in (8.3) corresponds to nonpropagating temperature (entropy) fluctuations and gives rise to the Rayleigh component (Landau and Placzek 1934) (these fluctuations propagate in superfluid and solid helium giving rise to second sound; see Section 8.3.1). To a good approximation the ratio of the intensity of the Rayleigh component to the sum of the two Brillouin components is

$$\frac{I_R}{2I_B}=\frac{C_P}{C_V}-1. \tag{8.4}$$

This is known as the Landau-Placzek ratio (Landau and Lifshitz 1960); it indicates that the intensity of Rayleigh scattering is proportional to C_P-C_V. One weakness of (8.4) arises from neglect of dispersion in thermodynamic properties, since the Brillouin components are measured at relatively high frequencies ($\gtrsim 10^9$ Hz) (Cummins and Gammon 1966).

The calculated line shapes for both Rayleigh and Brillouin components are Lorentzian (Mountain 1966a). For the Rayleigh component we get for the linewidth (Figure 8.1)

$$\Delta\omega_R=2\lambda' q^2, \tag{8.5a}$$

where $\lambda'=\lambda/\rho C_P$ is the thermal diffusivity, λ is the thermal conductivity and q is the wavevector transfer. In fluids the linewidth (8.5a) has a value of about 10 MHz for backscattering, decreasing as $\sin^2(\phi/2)$ as one goes to the forward direction, where ϕ is the scattering angle (Figure 8.2). The calculated width of the Brillouin components is (Figure 8.1)

$$\Delta\omega_B=2\alpha' q^2, \tag{8.5b}$$