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LASER LIGHT SCATTERING IN FLUID SYSTEMS

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Abstract

Recent theoretical and experimental advances in the field of inelastic light scattering from fluid systems are reviewed. Three basic frequency domains are distinguished by the

instrumentation used in each: (1) grating spectrometers, (2) optical interferometers, and (3) electronic spectrometers. Recent advances in each domain are discussed. A generalized hydrodynamic theory is outlined which incorporates both second-order and finite-frequency transport coefficient effects. Scattering experiments on density, anisotropy, and concentration fluctuations are discussed. Studies of surface waves, liquid crystals, intermolecular collisions, and fluid shear waves typify the kinds of experimental advances represented. Critical phenomena associated with various phase transitions in fluids and mixtures are brought up to date, and a discussion of light scattering as currently applied to macromolecular solutions is presented. The review concludes with a cross-referenced bibliography consisting of more than 500 titled entries on the subject of inelastic light scattering in fluid systems.

I. INTRODUCTION

Following the invention of the laser light source there has been an impressive increase in both the quantity and quality of optical scattering experiments performed on fluid systems of physical, chemical, and biological interest. Many of these experiments, particularly in the area of obtaining dynamical information, have been made possible by simultaneous advances in spectroscopic technique. Despite the relative newness of light scattering spectroscopy, several review articles on various aspects of the theory and experimental technique have already appeared. The article by McIntyre and Sengers¹⁰ presents an introduction to the basic concepts of light scattering in fluids and discusses at length the information that can be and has been obtained from such experiments regarding critical phenomena in both pure fluids and fluid mixtures. Mountain¹² has presented formal hydrodynamic theory pertaining to the aspects of dynamic liquid structure inferable from light scattering experiments. An article by Boon and Deguent³² has extended by techniques of generalized hydrodynamics the theory of light scattering in simple dense fluids. A thorough discussion of the techniques of electronic spectroscopy which utilize the coherence properties of the scattered field has been given by Cummins and Swinney.³

Of particular and growing interest to both chemists and biologists are the possible applications of laser scattering in fluids to probe macromolecular dynamics and chemical reaction kinetics. The theory of such experiments has been discussed in detail in two papers by Pecora.^{13,14} The experimental as well as theoretical aspects of light scattering in the study of chemical reactions have been reviewed by Yeh and Keeler.^{346,347} More wide ranging reviews of light scattering have been presented by Fleury,⁹ Chu,² and Benedek.^{1,217}

The purpose of this article is to review those recent developments not discussed in the above-mentioned works, and to place them in perspective relative to both the more familiar results and the areas showing promise

for future exploitation by light scattering spectroscopy. Detailed derivations of theoretical expressions are often omitted here. We concentrate rather on results, both theoretical and experimental. Further, some rather sweeping exclusions must be made at the outset regarding subject matter. We exclude the topics of ordinary Raman effect in fluids, of light scattering from solids and from plasmas, and of stimulated scattering of any kind. Each of these represents such a vast literature and so active a research field in its own right that a proper discussion of them in addition to our main subject would be impossible in a review article of reasonable length.

We begin in Section II with a discussion of theoretical generalities applicable to all types of scattering experiments. Results of the generalized hydrodynamic approach to the calculation of the density correlation functions, and their extension to multicomponent systems and to systems under conditions of nonequilibrium are presented. In addition orientational and relaxation effects on the spectra are examined in Section IV.

In Section III we emphasize recent improvements in experimental techniques and apparatus. These include multiple instruments (the tandem Fabry-Perot and other compound interferometers) on the one hand, and resonance reabsorption of the incident laser frequency, on the other hand. Basic considerations of electronic or "light beating" spectroscopy are reviewed, and recent advances in the theory and construction of digital correlators for collecting and processing data are discussed briefly as well.

Section IV concentrates on new types of experimental results. These include the study of phase transitions in liquid crystals, of intermolecular light scattering in simple fluids, of capillary waves at liquid interfaces, particularly in the vicinity of critical points, and of recent developments in both the theory and experimental observations on critical phenomena. Results in fluid mixtures and macromolecular solutions of interest to chemists and biologists are also discussed.

In the last section, a rather complete bibliography of the literature since the introduction of the laser to light scattering spectroscopy in the early 1960s is included. This bibliography divides the more than 500 citations (with titles included) into a dozen categories for hopefully easy access. To our knowledge it is the most complete compilation to date of the literature in this field, and we hope it will prove useful to both the neophyte and the seasoned research worker. With the exception of stimulated scattering, the subject exclusions mentioned above apply as well to the bibliography. The stimulated scattering citations revealed in our literature search are included here primarily because of their possible interest to readers.

The footnote and reference numbering procedure we adopt here is explained at the beginning of the section, and was dictated by the desire to include a complete bibliography as well as textual references with minimum redundancy and space expenditure.

As we see in the following section, when the kinematics of the scattering process are discussed more fully, the laser's monochromaticity and high degree of directional collimation have made the laser scattering technique valuable for the study of an incredible range of dynamic phenomena in fluids. Using a variety of spectroscopic techniques, fluctuations in the fluid occurring on time scales between 1 cps ranging in a continuous fashion up to 10^{16} cps (approximately the laser frequency itself) can be studied in a scattering experiment. By varying the scattering angle the spatial wavelength of the fluctuations probed can be selected over several orders of magnitude as well, ranging from a few thousand angstroms (for back scattering) to a few centimeters (for forward scattering). The practical upper limit is dictated by diffraction effects which exclude scattering angles smaller than the diffraction angle of the laser beam, $\sim \lambda/d$, where d is the laser beam diameter. This combination of dynamic range in both time and space, which makes light scattering such a powerful tool for the study of fluids, is shown in Fig. 1, where it is compared with those space-time regimes accessible to some other commonly used experimental techniques.

The light scattering domain is divided into three parts corresponding to generally different methods of spectrally analyzing the scattered light. The short-time or high-frequency phenomena occurring on time scales as short as 10^{-14} sec have been probed by means of grating spectrometers. This is the range of times involved, for example, in the intermolecular collision process, a very difficult region to probe by any experimental technique. In the intermediate range such familiar phenomena as Brillouin scattering from thermally excited acoustic waves and Rayleigh scattering from entropy fluctuations have received considerable attention. Similarly, the normal modes of vibration in complicated systems such as liquid crystals fall in this intermediate spectral range. These diverse phenomena have been studied conveniently with optical interferometers, having resolving powers as high as 10^9 .

The lowest energy region, corresponding to the highest resolution ($\sim 10^{15}$) has only recently emerged and owes its existence to the development of light beating and intensity correlation spectroscopy. Processes of interest in this region include the critical slowing down of density fluctuations near the liquid-gas phase transition, concentration fluctuations in critical mixtures, the diffusive motion of biological macromolecules in solution, and hydrodynamic instabilities in the vicinity of their incipient development.

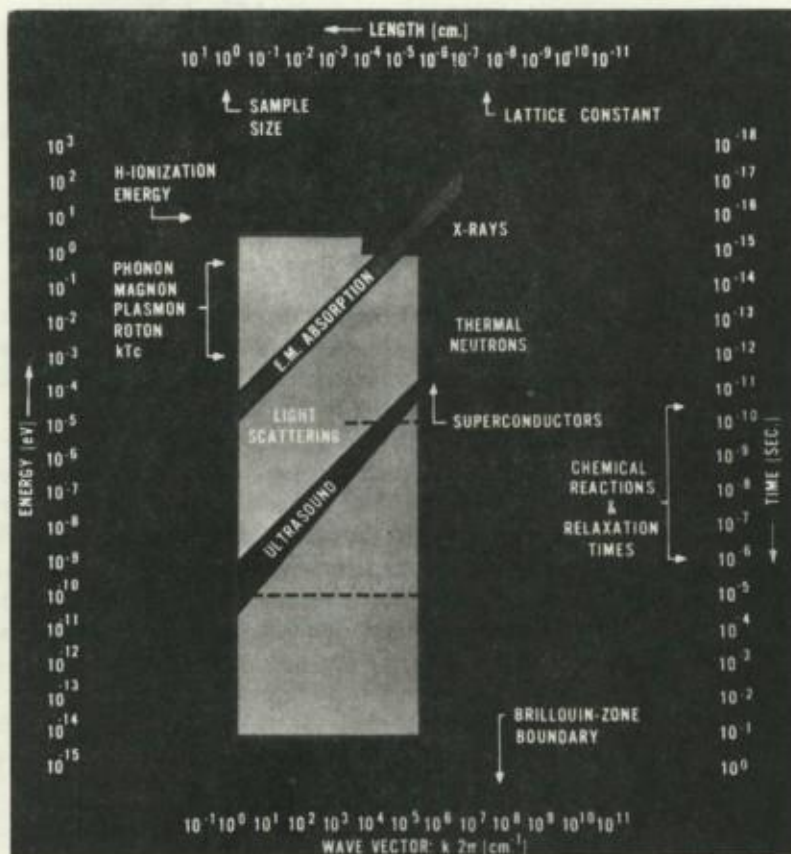


Fig. 1. Space-time or energy-momentum regimes accessible to various experimental techniques. Scattering techniques span rectangular areas by virtue of the additional kinematic range afforded by a pair of probe particles (incident and scattered). Note the logarithmic scales which permit display of large dynamic ranges.

The other scattering techniques depicted in Fig. 1 can probe shorter-wavelength fluctuations, but at present have not nearly the dynamic range in frequency available to light scattering.

II. THEORETICAL CONSIDERATIONS

A. General Relations

The scattering experiment is depicted schematically in Fig. 2, where an incident plane-polarized optical field E_1 , of frequency ω_1 , wave vector k_1 ,

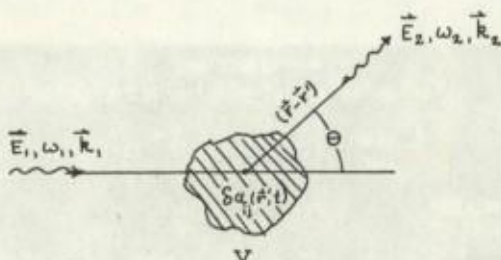


Fig. 2. Schematic diagram of scattering experiment.

and polarization $\hat{\epsilon}_1$ traverses a medium supporting time- and space-dependent polarizability tensor, $\delta\alpha_{ij}(\mathbf{r}', t)$. For a perfectly homogeneous polarizability, $\delta\alpha_{ij}(\mathbf{r}', t) = \text{Const.} \times \delta\alpha_{ij}(t)$, the reradiated (scattered) field would sum to zero for all but the exact forward direction. For a perfectly static polarizability, $\delta\alpha_{ij}(\mathbf{r}', t) = \text{Const.} \times \delta\alpha_{ij}(\mathbf{r}')$, the scattered light field would have the same frequency (time dependence) as incident field. The experiment generally consists of measuring the strength, polarization, frequency, and wave vector of the scattered field. These quantities carry information on the strength, symmetry, and time and spatial behavior of $\delta\alpha_{ij}$. The magnitude of $\delta\alpha_{ij}$ measures the scattering efficiency or cross section. Under the assumptions that (1) the scattering volume is much larger than λ_1^3 , (2) that the scattering volume dimensions are small compared to the distance between the scattering volume and the point of observation (\mathbf{r}) of the scattered field, and (3) that $\delta\alpha$ is sufficiently small that only a small fraction of the incident light is scattered, the scattered field may be written⁹:

$$E_2^{(i)}(\mathbf{r}, t) = \frac{-\omega_1^2 \sin \varphi}{|\mathbf{r} - \mathbf{r}'|^2 c^2} \eta_1^2 \exp(i\mathbf{k}' \cdot \mathbf{r} - \omega_1 t) \times \int_V dV' \exp(-i\mathbf{k} \cdot \mathbf{r}') E_1^{(j)} \delta\alpha_{ij}(\mathbf{r}', t) \quad (1)$$

where $(\eta_1 \omega_1 / c) |\mathbf{r} - \mathbf{r}'| = \mathbf{k}' \cdot (\mathbf{r} - \mathbf{r}')$ defines \mathbf{k}' and $\mathbf{k} \equiv \mathbf{k}' - \mathbf{k}_1$ and the superscript indexes the i th Cartesian component of \mathbf{E}_2 . φ is the angle between the incident polarization direction and the observation direction. Since the scattered intensity spectrum is simply related to the Fourier transform of the time correlation function of the scattered field, we have

$$I(\omega, \mathbf{r}) = \frac{c}{8\pi\mu} \left(\frac{\epsilon}{\mu}\right)^{1/2} \langle \mathbf{E}_2(\mathbf{r}, \omega_2) \cdot \mathbf{E}_2^*(\mathbf{r}, \omega_2) \rangle \quad (2)$$

where the angular brackets indicate an appropriate average over the states of the system. We see from (1) and (2) that the information about the

medium revealed by the scattering experiment is contained in the space-time correlation function of the polarizability tensor elements:

$$\langle \delta\alpha_{ij}(\mathbf{r}, t) \delta\alpha_{kl}^*(\mathbf{r} - \mathbf{r}', t - t') \rangle \quad (3)$$

Under assumption 1, above, the volume integral in (1) may be extended to infinity with the result that the scattered field at \mathbf{r} is strictly proportional to the q th Fourier component of $\delta\alpha_{ij}$. The time Fourier transform of \mathbf{E}_2 is just

$$\mathbf{E}_2(\omega_2) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathbf{E}_2(t) e^{i\omega_2 t} dt \quad (4)$$

and is proportional to $\delta\alpha_{ij}(\mathbf{k}, \Omega) e^{i(\omega_1 \pm \Omega)t}$. This reveals that (a) the Ω th Fourier component of $\delta\alpha_{ij}$ is observed and that (b) the scattered field frequency ω_2 is just $\omega_1 \pm \Omega$. Finally, the various polarization components of the scattered intensity are expressed by combining the above results as

$$I(\mathbf{r}, \omega_2) = \frac{V}{8\pi} \left(\frac{\epsilon}{\mu} \right)^{1/2} \frac{k_1^4 \sin^2 \varphi}{r^2 c} E_1^{(j)} E_1^{(i)} \langle \delta\alpha_{ij}(\mathbf{k}, \Omega) \delta\alpha_{ii}^*(\mathbf{k}, \Omega) \rangle \quad (5)$$

where summation on repeated indices is implied.

Implicit in this expression are the kinematics of the scattering process, which correspond to overall momentum and energy conservation in the scattering process.

$$\hbar\mathbf{k}_2 = \hbar\mathbf{k}_1 \pm \hbar\mathbf{k}$$

and

$$\hbar\omega_2 = \hbar\omega_1 \pm \hbar\Omega \quad (6)$$

The (-) and (+) signs correspond to gain and loss, respectively, of a quantum of energy $\hbar\Omega$ by the medium, and are called the Stokes and anti-Stokes components of the spectrum. For a system in thermal equilibrium the spectrum is symmetric about ω_1 when $\hbar\Omega \ll kT$, but more generally exhibits the relative demagnification by $e^{-\hbar\Omega/kT}$ on the anti-Stokes side required by the principle of detailed balance. The wave vectors in (6) form a triangle which for $\Omega \ll \omega_1$, ω_2 yields the useful relation between scattering angle θ and momentum transfer $\hbar k$:

$$\hbar k = 2\hbar k_1 \sin \frac{\theta}{2} \quad (7)$$

Thus variation of θ permits probing of fluctuations of wavelength $\lambda_1/2 < \lambda < d$, where λ_1 is the laser wavelength ($\sim 10^{-5}$ cm $^{-1}$) and d is the laser beam diameter, thereby defining the diffraction limit.

Equations (5) and (6) represent the fundamental relations for light scattering experiments. However, their very generality is indicative of a

lack of detailed physical content. The physical, chemical, and even biological processes which determine the structure and strength of $\delta\alpha_{ij}(\mathbf{r}, t)$ are so incredibly diverse that to write down $\delta\alpha_{ij}$ for a general fluid or fluid mixture would require a detailed microscopic theory of the dynamical aspects of the system affecting the polarizability. Because both single-particle and many-particle polarizabilities are sensitive to locations or configurations of the particles and groups, this means virtually *all* the dynamic aspects. Fortunately different processes contributing to $\delta\alpha_{ij}$ often occur on greatly different time or length scales and may vary differently with parameters like temperature, pressure, concentration, or molecular complexity. Their contributions can then often be separated and the dynamics of a particular process can be *directly* measured by the light scattering experiment.

For example, in a pure fluid of monatomic molecules, the dominant contribution to $\delta\alpha_{ij}$ comes from density fluctuations. These contribute only to diagonal elements of $\delta\alpha$ so that the scattered light has the same polarization as the incident light. The observed correlation function $\langle \delta\rho(-\mathbf{k}, 0) \delta\rho^*(\mathbf{k}, \omega) \rangle$ is simply the familiar dynamic structure factor $S(\mathbf{k}, \omega)$. Various models have been used to calculate S in various approximations: (a) the kinetic regime, single particles^{72,76}; (b) the hydrodynamic regime^{72,76,133}; and (c) the generalized hydrodynamic regime.³² When $k\Lambda < 1$ (Λ = particle mean free path), $S(\mathbf{k}, \omega)$ consists of a triplet of peaks centered at $\omega = 0$ and at $\omega = \pm qV_s$ arising from the nonpropagating (Rayleigh) and propagating (Brillouin) components of the density fluctuation at k .⁵⁷ These peaks are nearly Lorentzian in shape and of width $\Delta\omega_R = (\kappa/\rho C_p)k^2$ and $\Delta\omega_B = \alpha_s v_s/\pi$; where $\kappa/\rho C_p$ is the thermal diffusivity and α_s, v_s are the sound attenuation and velocity, respectively (see Fig. 3). Studies of the Rayleigh and/or Brillouin spectra were among the first applications of laser spectroscopy, and continue to be of interest because (a) the precise manner in which $\Delta\omega_R$ approaches zero near the liquid-gas critical point has important consequences for the understanding of critical phenomena, and (b) measurements of the Brillouin splitting, width, and shape reveal behavior of sound waves in a frequency range far above that of conventional ultrasonics (10^9 – 10^{10} Hz), and thus may reveal departures from classical hydrodynamics. The $S(\mathbf{k}, \omega)$ is even more interesting in fluids composed of polyatomic molecules, not only because vibrational relaxation frequencies often lie in the 10^9 – 10^{10} Hz range, but also because the structure of $S(\mathbf{k}, \omega)$ itself in a relaxing fluid is often qualitatively different from that in a simple fluid. In particular, a new central peak appears whose strength and shape are directly related to the strength and relaxation time of the energy exchange between density fluctuations and the internal degrees of freedom. Further interesting

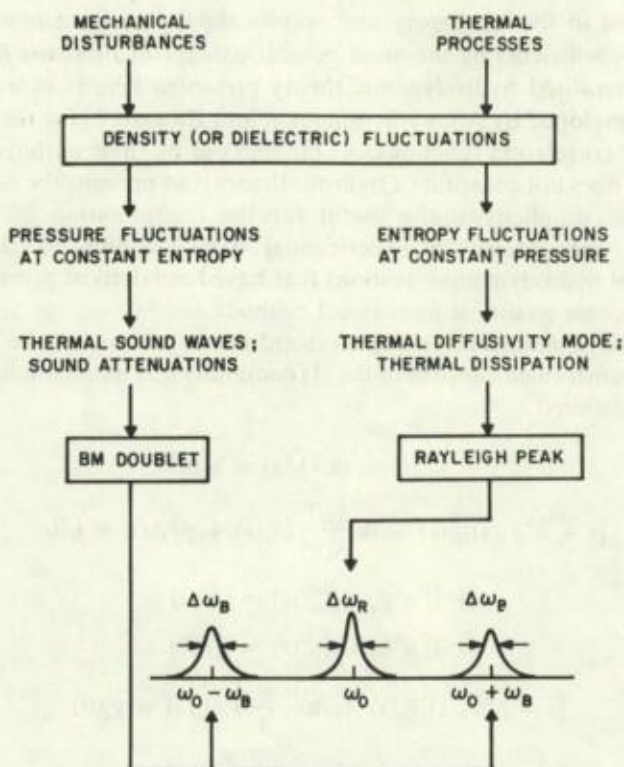


Fig. 3. Representation of the origin of the Rayleigh-Brillouin spectrum for a simple dense fluid.

effects on $S(\mathbf{k}, \omega)$ are expected in the generalized hydrodynamic regime ($\omega\tau_c \sim 1$, and $ka \sim 1$, with τ_c the collision time and a the characteristic intermolecular distance), where the ω and k dependence of thermodynamic and transport coefficients is taken into account. We consider here briefly the results of a description given by a generalized hydrodynamic description³² which unifies these contributions to $S(k, \omega)$.

B. Generalized Hydrodynamics for $S(\mathbf{k}, \omega)$

In this section, we first present a generalized hydrodynamics analysis as an extension of the hydrodynamic approach and then treat two particular examples as special cases, namely, the case of hypersound dispersion in simple liquids and the case of thermal relaxation resulting in the appearance of a new mode in liquids with internal degrees of freedom. Both cases correspond to actual experimental situations which have been

investigated in the laboratory and require the replacement of the usual transport coefficients by the more general concept of *transport functions*.

The generalized hydrodynamic theory presented here is based on the method developed by Ailawadi, Rahman, and Zwanzig⁵¹¹ for the analysis of current correlation functions. As pointed out by these authors, such a treatment does not constitute a rigorous theory, but presents the advantage of analytic simplicity—quite useful for the interpretation of physical situations encountered in experimental work—leading to the same generalized hydrodynamic equations that have been derived previously by more elaborate statistical mechanical methods.

The basic equations are straightforward generalizations of the Fourier-Laplace transformed versions of the (1) continuity, (2) momentum, and (3) energy equations:

$$s\tilde{\rho}_k(s) + i\mathbf{k} \cdot \tilde{\mathbf{j}}_k(s) = \rho_k(0) \quad (8)$$

$$[s + k^2\tilde{\varphi}_k(s)]\tilde{\mathbf{j}}_k(s) + i\mathbf{k} \frac{k_B T}{S(k)} [\tilde{\rho}_k(s) + \beta\tilde{g}_k(s)] = \mathbf{j}_k(0) \quad (9a)$$

$$[s + q^2\tilde{\varphi}_q(s)]\tilde{\mathbf{j}}_q^{(1)}(s) = \mathbf{j}_q^{(1)}(0) \quad (9b)$$

$$[s + q^2\tilde{\varphi}_q(s)]\tilde{\mathbf{j}}_q^{(2)}(s) = \mathbf{j}_q^{(2)}(0) \quad (9c)$$

$$[s + k^2\tilde{\Phi}_k(s)]\tilde{g}_k(s) + i\mathbf{k} \frac{\gamma - 1}{\beta} \cdot \tilde{\mathbf{j}}_k(s) = g_k(0) \quad (10)$$

where the quantities $\tilde{X}_k(s)$ are the Laplace-Fourier transforms of the fluctuations $\delta X(\mathbf{r}, t)$ defined as

$$\tilde{\rho}_k(s) = \int_0^\infty dt e^{-st} \int_V d\mathbf{r} e^{i\mathbf{k} \cdot \mathbf{r}} \delta\rho(\mathbf{r}, t) \quad (11)$$

with $s = \varepsilon + i\omega$, the complex Laplace variable. The longitudinal current \mathbf{j}_k and the transverse currents $\mathbf{j}_q^{(i)}$ ($i = 1, 2$) (with the wave vector \mathbf{q} in the plane perpendicular to the \mathbf{k} direction) have been defined as

$$\mathbf{j}_k(t) = \rho_0 \int_V d\mathbf{r} \mathbf{v}(t) e^{i\mathbf{k} \cdot \mathbf{r}} \quad (12)$$

$$\mathbf{j}_q(t) = \rho_0 \int_V d\mathbf{r} \mathbf{v}(t) e^{i\mathbf{q} \cdot \mathbf{r}} \quad (13)$$

Similarly, $g(\mathbf{r}, t)$ denotes the heat current density defined as

$$g(\mathbf{r}, t) = \rho_0 \delta T(\mathbf{r}, t) \quad (14)$$

In (8)–(10) we have employed the generalized coefficients defined as

$$\omega_0^2(k) = k^2 \frac{k_B T}{S(k)} = \frac{k^2}{\rho \chi_T(k)} \quad (15)$$

where $\chi_T(k)$ is the generalized isothermal compressibility

$$k^2 \bar{\varphi}_k(s) = [\omega_i^2(k) - \omega_0^2(k)] \bar{\psi}_k^l(s) \quad (16)$$

with

$$\omega_i^2(k) = \frac{\langle [\dot{j}_k(0)]^2 \rangle}{\langle [j_k(0)]^2 \rangle} \quad (17)$$

and where the index l denotes the longitudinal current;

$$k^2 \bar{\varphi}_q(s) = \omega_t^2(k) \bar{\psi}_q^l(s) \quad (18)$$

with

$$\omega_t^2(q) = \frac{\langle [\dot{j}_q(0)]^2 \rangle}{\langle [j_q(0)]^2 \rangle} \quad (19)$$

and where the index t denotes the transverse current;

$$k^2 \bar{\Phi}_k(s) = \omega_\lambda^2(k) \bar{\psi}_k^\lambda(s) \quad (20)$$

with

$$\omega_\lambda^2(k) = \frac{\langle [\dot{g}_k(0)]^2 \rangle}{\langle [g_k(0)]^2 \rangle} \quad (21)$$

In (16), (18), and (20), the functions $\bar{\psi}(s)$ are the memory functions of the master equations governing the time evolution of the corresponding currents; that is,

$$j(t) = - \int_0^t d\tau \psi(t - \tau) j(\tau) \quad (22)$$

An a priori calculation of these memory functions is quite difficult. Although the formal expressions for the ψ functions are analytically known, their explicit evaluation requires model calculations.

On the other hand, the ω 's can be computed from (17), (19), and (21), and the explicit expressions for the different currents, $j(0)$, and generalized forces, as well as some numerical computations, can each be found in the literature.

The set (8)–(10) constitute the generalized hydrodynamic equations. To make contact with classical hydrodynamics, it suffices to take the long-time limit,

$$\lim_{s \rightarrow 0} \bar{\psi}_k(s) = \tau(k) \quad (23)$$

where τ is the appropriate relaxation time for the transport process considered. Then one finds⁵¹¹

$$\lim_{k \rightarrow 0} [\omega_i^2(k) - \omega_0^2(k)]k^{-2}\tau_i = \nu_0 \quad (24)$$

$$\lim_{k \rightarrow 0} \omega_i^2(k)k^{-2}\tau_i = \nu_0' \quad (25)$$

$$\lim_{k \rightarrow 0} \omega_\lambda^2(k)k^{-2}\tau_\lambda = \lambda_0 \quad (26)$$

and

$$\lim_{k \rightarrow 0} \frac{k^2 k_B T}{S(k)} = k^2 \left(\frac{\partial \rho}{\partial p} \right)_T^{-1} = U_T^2 k^2 = \gamma^{-1} U_s^2 k^2 \quad (27)$$

where the usual quantities are denoted by $U_s = [(\partial p / \partial \rho)_s]^{1/2}$, the adiabatic sound velocity; $\gamma = C_p / C_v$, the ratio of the specific heats; and $\beta = -(\partial \rho / \partial T)_p / \rho_0$, the coefficient of thermal expansion. The transport coefficients, ν_0 , ν_0' , λ_0 , are the total kinematic viscosity, the shear kinematic viscosity and the thermometric conductivity, respectively:

$$\nu_0 = \frac{1}{\rho_0} \left(\frac{4}{3} \eta_s + \eta_B \right) \quad (28)$$

$$\nu_0' = \frac{\eta_s}{\rho_0} \quad (29)$$

$$\lambda_0 = \frac{\kappa}{\rho_0 C_v} = \gamma \lambda' \quad (30)$$

Different models may be constructed for the memory functions by assuming different mathematical forms for $\psi_k(t)$. These forms can be chosen and adjusted to fit the experimental data or may be computed from the sum rules. For the sake of simplicity, we restrict our attention here to the mathematically most convenient model, introducing the exponential ansatz

$$\tilde{\psi}_k^i(s) = [s + \tau_i^{-1}(k)]^{-1} \quad (31)$$

$$\tilde{\psi}_k^\lambda(s) = [s + \tau_\lambda^{-1}(k)]^{-1} \quad (32)$$

Here the relaxation times, $\tau_i(k)$ and $\tau_\lambda(k)$, should be considered as adjustable parameters to be determined, for example, by fitting the power spectrum to the molecular dynamics data.

However, when one is interested in the hydrodynamic regime, τ_i and τ_λ can be computed from the hydrodynamic limit equations, (24) and (26), combined with (17), (21), and (15) taken in the limit $k \rightarrow 0$. In the domain of light scattering, one may restrict oneself to the set of (8)–(10),

wherefrom one obtains a dispersion equation of the fifth degree. Indeed it is expected that two additional modes will arise from the introduction of the generalized transport coefficients when cast in the form of the exponential model, (31) and (32). To the lowest order in $\omega_i \tau_i$ ($i = l, \lambda$), the five roots of the dispersion equation read²²:

$$S_0 = -\frac{1}{\gamma} \omega_\lambda^2 \tau_\lambda \quad (\text{thermal diffusivity mode}) \quad (33)$$

$$S_\lambda = -\tau_\lambda^{-1} \quad (\text{thermal relaxation mode}) \quad (34)$$

$$S_l = -\tau_l^{-1} \quad (\text{viscous relaxation mode}) \quad (35)$$

$$S_\pm = \pm i\gamma^{1/2} \omega_0 - \Omega \quad (\text{Brillouin modes}) \quad (36)$$

with

$$\Omega = \frac{1}{2} \left[(\omega_l^2 - \omega_0^2) \tau_l + \left(1 - \frac{1}{\gamma} \right) \omega_\lambda^2 \tau_\lambda \right] \quad (37)$$

Note that the mode S_l could still be decomposed into a contribution from shear relaxation and a contribution from structural relaxation (related to the existence of the generalized bulk viscosity coefficient).

In a simple liquid the equations yield two additional shear modes

$$S_{i=1,2} = -v'q^2$$

which will not be considered in more detail here, because they have not been observed in simple liquids to date. Such modes would probably not be detectable by light scattering in simple isotropic liquids since there is no direct coupling between the transverse component of the velocity and the density fluctuations. However, in fluids with anisotropic molecules, such coupling does occur and has been invoked to explain the low-frequency depolarized spectrum observed in such cases. More will be said of these spectra in Section III.

In general, coupling occurs between these different modes [(33)–(36)] resulting in deviations from the simple Lorentzian shape of the classical spectral components. Such deviations are indications of the existence of dynamical structure in the system, which structure will then manifest itself as hypersonic dispersion, as modifications to the shape and the intensity of the spectral lines, as well as deviations from the classical Landau-Placzek ratio. Although in general these changes are expected to be small, several liquids have been studied experimentally, whose spectra could not be fully explained by the classical theory. In the next sections, we consider two examples of situations where the present treatment has been used for

the interpretation of (1) a new central component and (2) the possible existence of hypersound dispersion in simple liquids. Similar cases may occur also in fluid mixtures, as we shall see when considering the extension of the theory to multicomponent systems.

C. Internal Degrees of Freedom and Relaxation Processes

Energy transfer may occur between internal degrees of freedom and the translational degrees of freedom, in which case there will be coupling between a mode characteristic of structural relaxation and the collective modes describing the density fluctuations. This problem can be regarded as a particular case of the general prescription displayed in the previous section. There one restricts oneself to the limit $k \rightarrow 0$ and solely the bulk viscosity coefficient is generalized into the form of a transport function, with the following simple ansatz, assuming a single relaxation time process¹³¹ [see (16) and (31)]:

$$\begin{aligned} \lim_{k \rightarrow 0} k^2 \bar{\varphi}_k^B(s) &= \lim_{k \rightarrow 0} [\omega_B^2(k) - \omega_0^2(k)] \bar{\psi}_k^B(s) \\ &= \eta_B \frac{k^2}{\rho} + \frac{(U_\infty^2 - U_s^2) \tau_B k^2}{1 + s \tau_B} \end{aligned} \quad (38)$$

with U_∞ , the infinite frequency sound speed. Here the index B indicates that solely the contribution from the bulk viscosity is considered, and it has been found convenient to separate off the zero-frequency part of the function (i.e., the usual kinematic bulk viscosity coefficient, η_B/ρ_0). The dispersion equation can then be cast into the form

$$\left(s + \frac{\lambda k^2}{\gamma}\right)(s + i\omega_s + \Omega')(s - i\omega_s + \Omega') \left(s + \left(\frac{U_s}{v_s}\right)^2 \tau_B^{-1}\right) = 0 \quad (39a)$$

Here

$$\omega_s = v_s k = \frac{\tau_B^{-1}}{\sqrt{2}} [\Delta + \sqrt{\Delta^2 + 4(U_s k \tau_B)^2}]^{1/2} \quad (39b)$$

$$\Delta = [(U_\infty k \tau_B)^2 - 1]$$

$$\Omega' = \Omega + \frac{1}{2} \left[1 - \left(\frac{U_s}{v_s}\right)^2 \right] \frac{\lambda k^2}{\gamma} + \frac{1}{2} \frac{(U_\infty^2 - U_s^2) k^2}{\omega_s^2 + \tau_B^{-2}} (\tau_B^{-1} - \lambda k^2) \quad (39c)$$

Now from the dispersion equation (39), the spectral distribution can be obtained and may be *approximately* represented as a sum of *four* Lorentzian

curves¹³¹ (with $\Gamma_p = \lambda_0/\gamma$):

$$\begin{aligned} \frac{I(\omega)}{I_0} = & \left(1 - \frac{1}{\gamma}\right) \frac{2\Gamma_p k^2}{\omega^2 + (\Gamma_p k^2)^2} + \frac{2\left(\frac{U_s}{v_s}\right)^2 \tau_B^{-1}}{\omega^2 + \left(\frac{U_s}{v_s}\right)^4 \tau_B^{-2}} \\ & \times \frac{[1 - (v_s/U_s)^2][(1 - 1/\gamma)(U_s k)^2 + (U_s/v_s)^4 \tau_B^{-2}] + (U_\infty^2 - U_s^2)k^2}{\omega_s^2 + (U_s/v_s)^4 \tau_B^{-2}} \\ & + \frac{[1 - (U_s/v_s)^2(1 - 1/\gamma)][\omega_s^2 + (U_s/v_s)^2 \tau_B^{-2}] - (U_\infty^2 - U_s^2)k^2}{\omega_s^2 + (U_s/v_s)^4 \tau_B^{-2}} \\ & \times \left\{ \frac{\Omega'}{\Omega'^2 + (\omega + v_s k)^2} + \frac{\Omega'}{\Omega'^2 + (\omega - v_s k)^2} \right\} \end{aligned} \quad (40)$$

which reduces to the classical hydrodynamic spectrum when there is no dispersion ($U_s = U_\infty = v_s$). From the above spectrum, one obtains the ratio of the integrated intensity of the unshifted central components, $I_R^{(1)} + I_R^{(2)}$, to the integrated intensity of the Brillouin components to read

$$\frac{[I_R^{(1)} + I_R^{(2)}]}{2I_B} = (\gamma - 1) \frac{1 + A}{B} \quad (41)$$

where

$$\begin{aligned} A = & \frac{\gamma(U_\infty^2 - U_s^2)k^2 - [(v_s/U_s)^2 - 1] \left[\gamma \left(\frac{U_s}{v_s}\right)^4 \tau_B^{-2} + (kU_s)^2(\gamma - 1) \right]}{(\gamma - 1) \left[\omega_s^2 + \left(\frac{U_s}{v_s}\right)^4 \tau_B^{-2} \right]} \\ B = & \frac{[\gamma - (U_s/v_s)^2(\gamma - 1)][\omega_s^2 + (U_s/v_s)^2 \tau_B^{-2}] - \gamma k^2 (U_\infty^2 - U_s^2)}{\omega_s^2 + (U_s/v_s)^4 \tau_B^{-2}} \end{aligned}$$

which, at low phonon frequencies, that is, for $\omega_s \ll \tau_B^{-1}$, reduces to the classical Landau-Placzek ratio: $(\gamma - 1)$. In the limit of high frequencies, that is, $\omega_s \gg \tau_B^{-1}$, one retrieves Rytov's result⁶⁶

$$(\gamma - 1) \left\{ 1 + \frac{\gamma}{\gamma - 1} \frac{U_\infty^2 - U_s^2}{U_s^2} \right\} \quad (42)$$

The most striking feature of the spectrum given by (40) is, of course, the existence of a second central component due to structural relaxation with

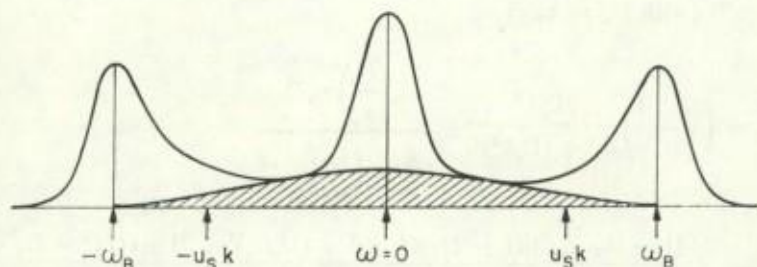


Fig. 4. Spectrum of density fluctuations for a singly relaxing fluid. The relaxation central component (shaded area) causes the Brillouin peaks (at $\pm\omega_B$) to be displaced from their zero-order positions (at $\pm u_s k$).

relaxation time τ_B . The spectral distribution is illustrated in Fig. 4, where the shaded area represents the new spectral component, which has been referred to as the "Mountain peak." Actually, in most experimental spectra, the existence of such a relaxation mode will manifest itself as a broad background between the Rayleigh component and the Brillouin peaks, which in turn will appear asymmetrical and will also be displaced from their classical position (at $\omega = U_s k$) to a new position, $\omega_s = v_s k$ according to (39a) and (39b). Notice, however, that ω_s/k measures the propagation velocity, whereas the maximum of the Brillouin component occurs at ω_B , for which $\partial I(\omega)/\partial\omega = 0$, and because of the asymmetrical terms [see (57)] in the spectral distribution, one has $\omega_B < \omega_s$. On the other hand, the effect of the broad central component (due to structure relaxation) results in a pushing of the Brillouin peaks away from the Rayleigh line, that is, $\omega_s > u_s k$. Thus considerable care must be taken to extract the sound velocity from the Brillouin spectrum in a relaxing fluid.

The present method can of course be extended to multiply relaxing systems. The case of both singly and doubly relaxing liquids has been investigated by Nichols and Carome.¹³⁴ In the latter case, a second relaxation time was assigned to the generalized longitudinal viscosity, with the assumption of an exponential ansatz.

D. Transport Function and High-Frequency Effects

In this section, we shall consider the case of simple monatomic liquids, uncomplicated by the relaxation processes associated with internal degrees of freedom. Even so the question of the possible frequency dependence of the transport coefficients in the "intermediate domain" characterized here as $k \rightarrow 0$, $\omega/\omega_c \sim 10^{-2}$ becomes of interest. ω_c is a characteristic frequency, of order of the collision frequency above which translational

relaxation effects are important. The "intermediate domain" thus lies between the hydrodynamic domain ($k \rightarrow 0$, $\omega \rightarrow 0$) and the generalized hydrodynamic domain ($ka \sim 1$, $\omega\tau_c \sim 1$). Indeed it has been shown in transport theory that the linear response of a classical fluid becomes wholly nondissipative in the very high frequency limit; that is, when $\omega \rightarrow \infty$, the transport coefficients become purely imaginary.¹¹⁰ We recall that in general, any transport coefficient at finite frequency, that is, any *transport function*, may be cast into a correlation expression as⁵⁰⁵

$$\sigma(\omega) = c \int_0^{\infty} dt e^{i\omega t} \langle J(0)J(t) \rangle \quad (43)$$

where c is a thermodynamic constant and $J(t)$ is the flux related to the transport property considered. As a result, the transport function is generally complex

$$\sigma(\omega) = \sigma_R(\omega) + i\sigma_I(\omega) \quad (44)$$

The zero frequency term, $\sigma_R(0)$, is merely the usual transport coefficient, and the first term in the ω expansion appears to be imaginary, constituting the first nondissipative contribution at high frequencies. Because for light scattering the accessible ω values place us in the intermediate domain, we may legitimately restrict ourselves to the lowest order in ω , in which case the transport functions take the form

$$\nu(\omega) = \lim_{\substack{k \rightarrow 0 \\ \epsilon \rightarrow 0}} \tilde{\Phi}_k(s) = \nu_0(1 - i\alpha_1\omega) \quad (45)$$

$$\lambda(\omega) = \lim_{\substack{k \rightarrow 0 \\ \epsilon \rightarrow 0}} \tilde{\varphi}_k(s) = \lambda_0(1 - i\alpha_2\omega) \quad (46)$$

where ν_0 and λ_0 are the zero frequency transport coefficients as given by (28) and (30). α_1 and α_2 are of the order of ω_c^{-1} and can be calculated explicitly [see (17) and (21)]; for example, one finds indeed that for the shear viscosity, $\alpha\omega \simeq 10^{-2}$. With these assumptions the dispersion equation must now be solved to second order in quantities like $\lambda k/U_s \equiv a$ and $\nu k/U_s \equiv b$, because, although these second-order terms are small, they are no longer negligible with respect to the $\alpha\omega$ terms in (45) and (46). The modes are now given by³²

$$s_0' = -\Gamma_p' k^2 = -\Gamma_p k^2(1 - \delta_1) \quad (47)$$

$$s_{\pm}' = \pm i\nu_s k - \Gamma_s' k^2 = \pm iU_s k(1 - \delta_2) - \Gamma_s k^2(1 - \delta_3) \quad (48)$$

where Γ_p and Γ_s are given by $\Gamma_p = \lambda_0/\gamma$; $\Gamma_s = v_0 + (\gamma - 1)\lambda_0/\gamma$ [see (28) and (30)],

$$\delta_1 = \left(1 - \frac{1}{\gamma}\right) \left(\frac{b}{a} - \frac{1}{\gamma}\right) a^2 \quad (49)$$

$$\delta_2 = \delta_2^{(\omega)} + \delta_2^{(2)} \quad (50)$$

$$\delta_2^{(\omega)} = -\frac{1}{2} \left[\left(1 - \frac{1}{\gamma}\right) \mu_\lambda a + \mu_\ell b \right] \quad (51)$$

$$\delta_2^{(2)} = \frac{1}{2} [\tilde{\lambda}^2 - \delta_1] \quad (52)$$

$$\delta_3 = \delta_3^{(\omega)} + \delta_3^{(2)} \quad (53)$$

$$\delta_3^{(\omega)} = \frac{a\mu_\lambda}{\gamma} + \delta_2^{(\omega)} \left(1 + \frac{a}{\tilde{\lambda}\gamma}\right) \quad (54)$$

$$\delta_3^{(2)} = \frac{1}{\gamma} \left(\frac{1}{\gamma} - \frac{b}{2\tilde{\lambda}}\right) a^2 \quad (55)$$

with the reduced quantities

$$\tilde{\lambda} = \frac{\Gamma_s k}{U_s}; \quad \mu_i = \alpha_i U_s k (i = \lambda, \ell) \quad (56)$$

The "high-frequency" effects arising from the nondissipative part of the transport functions have been separated off and appear in those terms with superscript (ω), whereas the other correction terms, labeled with superscript (2), represent pure second-order effects. Notice that the latter are the only corrections which affect the diffusion mode, whereas both the frequency and the lifetime of the thermal phonons are modified by the frequency effects.

With the correction terms discussed above, the spectrum of the scattered light reads³²

$$\begin{aligned} \frac{I(\omega)}{I_0} = & \left(1 - \frac{1}{\gamma}\right) (1 + \theta_1) \frac{2\Gamma_p' k^2}{\omega^2 + (\Gamma_p' k^2)^2} \\ & + \frac{1}{\gamma} (1 + \theta_2) \left\{ \frac{\Gamma_s' k^2}{(\omega + v_s k)^2 + (\Gamma_s' k^2)^2} + \frac{\Gamma_s' k^2}{(\omega - v_s k)^2 + (\Gamma_s' k^2)^2} \right\} \\ & + \frac{1}{\gamma} \theta_3 \left\{ \frac{\omega + v_s k}{(\omega + v_s k)^2 + (\Gamma_s' k^2)^2} - \frac{\omega - v_s k}{(\omega - v_s k)^2 + (\Gamma_s' k^2)^2} \right\} \quad (57) \end{aligned}$$

and a modification of the Brillouin shift inducing hypersound dispersion, that is,

$$\omega_B' = \pm U_s k(1 - \delta_2)(1 - \varepsilon) \quad (64)$$

with

$$\varepsilon = \frac{3\theta_2^2 + \theta_3^2}{2\theta_3} \frac{1 - \delta_3}{1 - \delta_2} \bar{\lambda} \quad (65)$$

Now (64) may be rewritten as

$$v_s \simeq U_s(1 - \delta_2^{(\omega)} - \varepsilon^{(2)}) \quad (66)$$

with

$$\delta^{(2)} + \varepsilon = \varepsilon^{(2)} \quad (67)$$

so as to separate the effects of the second-order terms, $\varepsilon^{(2)}$, from pure dispersion effects arising from the high-frequency corrections, $\delta_2^{(\omega)}$. Since the α 's ($\simeq \tau_\alpha$) are positive, $\delta_2^{(\omega)} < 0$, which as a consequence induces positive dispersion. Conversely, the effects due to second-order correction, with $\varepsilon^{(2)} > 0$, manifest themselves as a negative dispersion. The importance of these effects for the interpretation of Brillouin spectra observed in simple monatomic liquids like argon and neon are mentioned in Section III.

E. Light Scattering and Ultrasonics

Brillouin scattering may appear as a logical extension of ultrasonics to probe excitations in fluid systems by extending the domain of accessible frequencies to the GHz range. Such extension of course involves the problems discussed in this section, like dispersion effects, in particular for systems exhibiting relaxation processes, as is the case, for example, when internal degrees of freedom play an important role. Care should be taken, however, in the interpretation of light scattering results as compared to ultrasonic data, because of the boundary conditions which are not necessarily equivalent in both cases. Indeed when solving the dispersion equation, attention must be paid to the fact that either of two different boundary conditions may be appropriate. (1) When the frequency of the excitation is real and fixed by the experimental conditions, one faces the phenomenon of spatial absorption, commonly encountered in most ultrasonics experiments, whereas (2) temporal absorption occurs when the experimental conditions fix the wave vector, as in light scattering experiments.^{102,131}

It was predicted in the previous section that the phase velocity measured in a light scattering experiment, for a system without structural or thermal relaxation, should exhibit negative dispersion. To compare this result with the prediction for the reciprocal situation of a spatially damped wave, we may, for the sake of clarity, consider the simplified case of a system

with zero thermal conductivity; then the dispersion equation reduces to

$$s^2 + \nu k^2 s + (U_0 k)^2 = 0 \quad (68)$$

where U_0 denotes the zero frequency and infinite wavelength limit of the sound velocity. For k real, and $s = i\omega + \varepsilon$, the solution of (68) reads

$$s = \pm i\nu_s k - \frac{1}{2}\nu k^2 \quad (69)$$

with the hypersonic velocity

$$\nu_s = \pm U_0 \left[1 - \frac{1}{4} \left(\frac{\nu k^2}{U_0 k} \right)^2 \right]^{1/2} \quad (70)$$

(here we use the notation ν_s for the hypersound velocity, and U_s for the ultrasonic velocity) exhibiting indeed negative dispersion. On the other hand, for $s = i\omega$ (real frequency) and $k = k' + ik''$, the dispersion equation yields the set

$$k'^2 - k''^2 = \left(\frac{\omega}{U_0} \right)^2 \bar{U}_0^{-1} \quad (71)$$

$$2k'k'' = - \left(\frac{\omega}{U_0} \right)^2 \frac{\nu}{u_0} \bar{U}_0^{-1} \quad (72)$$

with

$$\bar{U}_0 = 1 + \left(\frac{\nu\omega}{U_0^2} \right)^2 \quad (73)$$

from which one obtains the "ultrasonic" velocity:

$$U_s = \frac{\omega}{k'} = \pm U_0 \left[\frac{2\bar{U}_0}{1 + \bar{U}_0^{1/2}} \right]^{1/2} \quad (74)$$

The latter, for low frequencies, that is, $\omega \ll (uk)^2/\nu k^2$, may be expanded to yield

$$U_s \simeq \pm U_0 \left[1 + \frac{3}{4} \left(\frac{\nu\omega}{U_0^2} \right)^2 \right]^{1/2} \quad (75)$$

from which it is clear that for complex k the dispersion is positive.

From (70) and (75) one observes that ν_s is always smaller than U_s except for the limiting case $k \rightarrow 0$, $\omega \rightarrow 0$, for which one finds the expected equality $\nu_s(k \rightarrow 0) = U_s(\omega \rightarrow 0) = U_0$.

Considering next the case of a relaxing system, one may introduce a generalized bulk viscosity to account for thermal relaxation. Such an analysis for the comparison of ultrasonics with light scattering can be performed along the same lines as for the simple case treated above; we

merely quote here the results of the analysis for the sound velocity in order to exhibit the dispersion effects.¹⁰²

$$v_s = \pm \frac{U_0}{\sqrt{2}} \left[1 + \frac{\omega \eta_2''(\omega)}{\rho_0 U_0^2} + \frac{|c|^2}{U_0^2} \right]^{1/2} \quad (76)$$

$$U_s = \pm \sqrt{2} \frac{|c|^2}{U_0} \left[1 + \frac{\omega \eta_2''(\omega)}{\rho_0 U_0^2} + \frac{|c|^2}{U_0^2} \right]^{-1/2} \quad (77)$$

with

$$\frac{|c|^2}{U_0^2} = \left\{ \left[1 + \frac{\omega \eta_2''(\omega)}{\rho_0 U_0^2} \right]^2 + \left[\frac{\omega \eta_2'(\omega)}{\rho_0 U_0^2} \right]^2 \right\}^{1/2} \quad (78)$$

Here, $\eta_2'(\omega)$ and $\eta_2''(\omega)$ are the real and imaginary parts of the second viscosity, respectively. Using, for example, a single relaxation time model, for $\eta_2(\omega)$, with relaxation time τ_2 , one has

$$\eta_2(\omega) = \frac{(U_\infty^2 - U_0^2)\tau_2}{1 + i\omega\tau_2} \rho_0 = \eta_2' + i\eta_2'' \quad (79)$$

It is then easily verified that in the limit $\omega \rightarrow \infty$, one obtains $v_s = U_s = U_\infty$ (whereas for $\omega \rightarrow 0$, of course, $v_s = U_s = U_0$). For the intermediate-frequency range, that is, when $\omega\tau \sim 1$, the use of a simple model permits the explicit computation as well as the numerical comparison of the dispersion effects. Probably the most interesting feature is that it is clear from (76) and (77) that relaxation effects induce positive dispersion for both U_s and v_s . Furthermore, here again one observes that one has always $U_s > v_s$, except in the limits discussed above, for which both the ultrasonic velocity and the hypersonic velocity tend to the same asymptotic values.

F. Solutions and Noncritical Mixtures

The spectra of multicomponent fluids are much more complex than for pure fluids. Advantage has been taken of this additional spectral information to measure mass diffusion coefficients,³⁵⁶⁻³⁵⁹ chemical reaction rates,^{346,347} departures from solution ideality,³¹⁰ etc., in solutions and binary liquid mixtures at thermal equilibrium. Although no experiments have yet been reported, recent theoretical discussions on fluids and mixtures *not* in equilibrium have suggested this as a fruitful area for future investigations. We present here in brief summary some of the principal theoretical predictions for both cases.

Mountain and Deutch³⁵⁰ have calculated the spectrum of the coupled pressure, (p), entropy, (φ), and concentration (c) fluctuations in a binary fluid mixture at equilibrium. For their hydrodynamic analysis one must

add to the basic conservation equations (8)–(10), the diffusion equation:

$$\frac{\partial c}{\partial t} = D \left[\nabla^2 c + \frac{k_T}{T} \nabla^2 T + \frac{k_p}{P} \nabla^2 p \right] \quad (80)$$

where c is the minority concentration, D the mass diffusion coefficient, k_T the thermal diffusion ratio = $c(1-c)(D'/D)$ (D' = thermal diffusion coefficient), and $k_p = c(1-c)(D''/D)p$, in terms of the baro-diffusion coefficient, D'' . The resulting spectrum $S(\mathbf{k}, \omega)$ is³⁰

$$\begin{aligned} & \left(\frac{\partial \alpha}{\partial p} \right)_{\varphi, c}^2 \frac{k_B T \rho}{\beta_s} \left\{ \frac{\Gamma k^2}{(\Gamma k^2)^2 + (\omega + U_0 k)^2} + \frac{\Gamma k^2}{(\Gamma k^2)^2 + (\omega - U_0 k)^2} \right\} \\ & + \left(\frac{\partial \alpha}{\partial \varphi} \right)_{c, p}^2 \left(\frac{k_B T}{C_p} \right) \left(\frac{2}{s_2 - s_1} \right) \left\{ \frac{(Dk^2 - s_1)s_1}{s_1^2 + \omega^2} - \frac{(Dk^2 - s_2)s_2}{s_2^2 + \omega^2} \right\} \\ & + \left(\frac{\partial \alpha}{\partial c} \right)_{\varphi, p}^2 \left(\frac{k_B T}{(\partial \mu / \partial c)_{p, T}} \right) \left(\frac{2}{s_2 - s_1} \right) \left\{ \frac{(s_2 - Dk^2)s_1}{s_1^2 + \omega^2} + \frac{(s_1 - Dk^2)s_2}{s_2^2 + \omega^2} \right\} \\ & + \left(\frac{\partial \alpha}{\partial \varphi} \right)_{p, c} \left(\frac{\partial \alpha}{\partial c} \right)_{p, \varphi} \left(\frac{2k_B T}{C_p} \right) \left(\frac{s_1}{s_1^2 + \omega^2} - \frac{s_2}{s_2^2 + \omega^2} \right) \left(\frac{2Dk^2 k_T}{s_1 - s_2} \right) \quad (81) \end{aligned}$$

where Γk^2 is the Brillouin width:

$$\Gamma = \Gamma_0 + \frac{DU_0^2}{\rho(\partial \mu / \partial c)_{p, T}} \left\{ \left(\frac{\partial \rho}{\partial c} \right)_{p, T} + \frac{k_T}{C_p} \left(\frac{\partial \rho}{\partial T} \right)_{p, c} \left(\frac{\partial \mu}{\partial c} \right)_{p, T} \right\}^2 \quad (82)$$

$\Gamma_0 = v_0 + (\gamma - 1)\lambda_0$ from (28) and (30). The roots s_1 and s_2 refer to the central components and are

$$s_1 = \frac{1}{2}\Lambda k^2 + \frac{1}{2}[\Lambda^2 k^4 - 4\lambda_0 D k^4]^{1/2} \quad (83)$$

$$s_2 = \frac{1}{2}\Lambda k^2 - \frac{1}{2}[\Lambda^2 k^4 - 4\lambda_0 D k^4]^{1/2} \quad (84)$$

$$\Lambda \equiv \lambda_0 + \mathcal{D} \quad (85)$$

$$\mathcal{D} = D \left[1 + \frac{k_T^2}{TC_p} \left(\frac{\partial \mu}{\partial c} \right)_{p, T} \right] \quad (86)$$

Here μ is the chemical potential.

The spectrum (81) contains in addition to the two propagating Brillouin modes, whose line width Γ is slightly modified, a complicated central feature due to the coupled entropy-concentration fluctuations. Only under certain simplifying conditions can this be decomposed into two Lorentzians corresponding to the thermal diffusivity and mass diffusion processes. In the very dilute limit, $c \ll 1$, we have $k_T \rightarrow 0$ and the two Lorentzians emerge with $s_1 \rightarrow \lambda_0 k^2 / \gamma$, $s_2 \rightarrow -Dk^2$. For small but finite c these peaks are quasi-Lorentzian with somewhat modified line widths. In particular the thermal diffusivity (Rayleigh) peak is broadened by an amount

$k^2(k_T/TC_p)(\partial\mu/\partial c)_{p,T}$, whereas the diffusion (concentration) peak is narrowed by $k^2(\partial\mu/\partial c)_{p,T}D(k_T/TC_p\lambda_0)$. Although several experiments on fluid mixtures of low c have been reported,^{28,310} the central spectra have been interpreted as two Lorentzians. Thus neither the Brillouin line width modifications nor the central line shape modifications discussed here have yet been measured for liquids. However, recent work of Gornall et al.³⁰⁸ has revealed striking interaction effects in disparate gas mixtures. And Lekkerkerker and Boon⁵¹² have reinterpreted the coupling effects observed in such mixtures.

Recently Boon²⁹⁷ has extended these considerations to include the very interesting *nonequilibrium* case of a horizontal binary fluid layer subjected to an adverse vertical temperature gradient. When a fluid layer is heated from below, the system becomes topheavy and potentially unstable (provided that the fluid has a positive thermal expansion coefficient). Therefore the fluid has a tendency to redistribute itself. This natural tendency is inhibited, however, by the viscosity of the fluid and by its thermal conductivity which seeks to relieve the imposed gradient. As a consequence the temperature gradient must exceed a certain value before the instability can manifest itself. When the temperature gradient reaches this critical value (depending on the properties of the fluid and on the thickness of the layer) convective motion begins which permits the fluid to adjust. Convection arises in a peculiar way, however, in that a stationary instability occurs, which results in an arrangement of spatially ordered convection cells or rolls, first demonstrated by Bénard in 1900.⁵¹⁰

In many ways the onset of this hydrodynamic instability, although a manifestly nonequilibrium phenomenon, bears close analogies to several well-studied second-order equilibrium phase transitions. Particularly intriguing is the behavior of fluctuations very near the instability point. Although in principle amenable to light scattering examination, the Bénard instability presents tremendous experimental challenges and no successful dynamic experiments have yet been reported. Nevertheless, Boon's theoretical predictions deserve summary here. He considers the dilute fluid mixture ($c \ll 1$) to form a layer of thickness, d , between two free boundaries and to be subjected to a temperature gradient $\beta = (T_h - T_c)/d$. β is conveniently expressed in dimensionless form as the Rayleigh number:

$$R = \frac{\alpha\beta g}{\lambda_0\nu_0} d^4 \quad (87)$$

where α is the thermal expansion coefficient and g the gravitational acceleration. The instability occurs for a particular value of R , called R_c , which depends only on the fluid layer boundary conditions. Boon has

examined the behavior of both the Rayleigh and concentration peaks as $R \rightarrow R_c$. He finds that the Rayleigh peak is most strongly affected when the spectrum (for a very small range of k_{\perp} 's near $k_c = \pi/2d$) approaches zero line width and infinite peak height as $R \rightarrow R_c$. In particular, the Rayleigh width is $(\lambda_0 k^2 / \gamma) [(1 - R/R_c)/(1 + 2R/R_c)]^{1/2}$ for a pure fluid.^{297,512} For a mixture near the instability point, the concentration mode is narrowed and the Rayleigh peak broadened slightly.

The application of light scattering to the study of hydrodynamic instabilities has thus far been rather limited. However, the recent experiments by Bertolotti et al.²⁷² on electric field induced instabilities in liquid crystals suggest how important a role light scattering can be expected to play in examining the dynamical aspects of nonequilibrium phenomena in fluids.

III. RECENT ADVANCES IN EXPERIMENTAL TECHNIQUE AND APPARATUS

Since the physically interesting phenomena amenable to examination by light scattering span so broad a range of characteristic lengths (10^{-5} –1 cm) and times (~ 1 – 10^{-15} sec) and exhibit such a wide variety of scattering efficiencies (10^{-1} – 10^{-12} $\text{cm}^{-1}\text{-Sr}^{-1}$) the experimental problems encountered are manifold indeed. In its most general form the basic experimental problem consists in measuring the spectral content of the usually weak inelastically scattered light in the presence of a much stronger signal arising from elastic or parasitic scattering. One is often faced with the problem of simultaneously optimizing the spectral resolution and contrast of the detection system in a manner determined by the strength and time domain of the fluctuations to be studied. The spectral domain can be conveniently divided into the three areas, delineated above with regard to Fig. 1. In this section we present a brief description of experimental techniques and recent advances therein applicable to each frequency range.

Although some interesting progress has been made in the area of laser sources (such as tunable dye lasers, the He–Cd laser, and ultrashort pulse lasers) in recent years, the great majority of laser spectroscopy in fluids continue to utilize the He–Ne or argon ion laser. Single-frequency stabilized versions of both lasers are now commercially available, so no further discussion of sources is required here.

A. High-Frequency Shifts

Typical meter-long grating spectrometers are routinely capable of studying frequency shifts in the 0.2 to ≥ 2000 cm^{-1} range (a maximum resolving power of $\sim 10^6$). Double-grating instruments consisting of two such spectrometers in series are available which increase the contrast to

$\sim 10^{10}$ with virtually no sacrifice in resolving power. Such spectrometers have long been used for studies of rotational and vibrational Raman spectra in liquids, gases, and solids. However, some fluid phenomena of recent interest lie in this frequency range and their study has been carried out using grating spectrometers. Both anisotropy scattering from fluctuations in molecular orientation and intermolecular or "collision-induced" scattering fall into this category. The latter is closely related to the recently observed but very weak scattering of light by pairs of rotons in superfluid helium. About the only recent instrumental improvement in grating spectrometers has been the addition of a third stage—usually a lower-resolution premonochromator—to further enhance the contrast while leaving the resolution unaffected. Primarily of importance for solids where parasitic scattering is much more severe than in fluids, such improvements will not be discussed further here.

B. Intermediate-Frequency Shifts

The second spectral area denoted in Fig. 1 lies between 10^6 Hz and 10^{11} Hz (10^{-4} – 1 cm^{-1}) and necessitates the use of optical interferometers. By far the most widely used is the Fabry-Perot (FP) interferometer, which in its simplest embodiment consists of a sharp tunable interference filter. Complete discussions of its operation are available in a variety of references.^{9,1009} There are two basic versions generally employed for fluid experiments, the flat FP and the spherical FP. The former consists of two flat plane-parallel low-loss mirrors of reflectivity R separated by an optical path length nd (n = refractive index of the medium separating the plates). The resolving characteristics of the FP are summarized⁹ by the fraction of light I_p/I_0 traveling parallel to the interplate axis, passed at frequency ν :

$$\frac{I_p}{I_0} = \left[1 + \frac{4R}{(1-R)^2} \sin^2 \left(2\pi\nu \frac{nd}{c} \right) \right]^{-1} \quad (88)$$

The transmission is periodic in $\Delta\nu = 2c/nd \equiv$ free spectral range $\equiv f$. When the \sin^2 term equals unity, the minimum I_p/I_0 obtains and is $\approx (1-R)^2/4R$, which determines the contrast. For $R = 0.98$ the contrast for a single FP is $\sim 10^4$. The resolving power depends on the free spectral range and on the "finesse," which is the ratio of f to $\delta\nu$. $\delta\nu$ is the full frequency width at half maximum of the transmission peaks in the function (1). Typical values of finesse are < 100 . Thus for an f of 100 MHz the resolving power may approach 10^9 . This value of f requires a plate separation d of order 150 cm. More typical for fluid studies are 1–10 GHz for spectral ranges corresponding to conveniently smaller values of d . Scanning of the pass frequency of the flat FP is most often achieved by varying the

gas pressures (and hence n) between the interferometer plates. This is desirable because no physical motion of the plates is required which could easily misalign the optical cavity and degrade the instrument's spectral characteristics.

The incipient instability of the flat optical cavity can be obviated by using instead a spherical FP, two identical spherical mirrors separated by a distance r (r = radius of curvature). This confocal configuration is much more stable and permits reliable scanning of the FP by slight physical modulation of the mirror separation, as well as by the pressure variation previously mentioned. The light gathering properties of the spherical FP are superior to those of the flat FP for large r . The main disadvantage lies in the lack of adjustable f for the spherical FP. Finesse and resolving power considerations are quite similar to those for the flat FP.⁹

For many fluid applications either of these simple FP's may be quite adequate. However, some situations arise (such as diverging Rayleigh intensity near the critical point, or quasielastic scattering from particles in suspension) where a greater contrast than 10^4 is required. There are two ways in which this has been achieved: first, utilization of compound interferometers; and second, resonant reabsorption of the elastically scattered light.

1. Compound Interferometers

The most obvious direction for improvement by this method is the simple series arrangement of two or more FP's. This indeed has been discussed in some detail by Mack et al. nearly ten years ago.²⁵ Problems with simultaneity of scan, intercavity resonances, and overall instrumental line shape are of course considerably more complicated than with the single FP. Three different types of compound interferometers have been demonstrated and deserve mention here. In 1969 Langley and Ford²² introduced in front of their FP a narrow prefilter consisting of an unequal-arm Michelson interferometer to prevent over 99% of the elastically scattered light from reaching the FP. Since the Michelson is not scanned, the system behaves very much like a simple FP with an enhanced contrast ($\sim 10^6$) and a more complicated instrumental function.

Cannell²⁰ has operated a tandem spherical FP interferometer for his Brillouin experiments near the critical point in Xe. Two FP's of differing f 's ($f_1 = 1510$ MHz, $f_2 = 991$ MHz) were placed in series inside a common pressure cell to ensure perfect tracking during the frequency scan. One FP incorporated a piezoelectrically tunable offset to allow initial simultaneous tuning of both FP's for peak transmission. The instrumental transmissions of each component as well as the compound instrument are shown in Fig. 6. The contrast is thus essentially squared ($\sim 3 \times 10^6$) and the

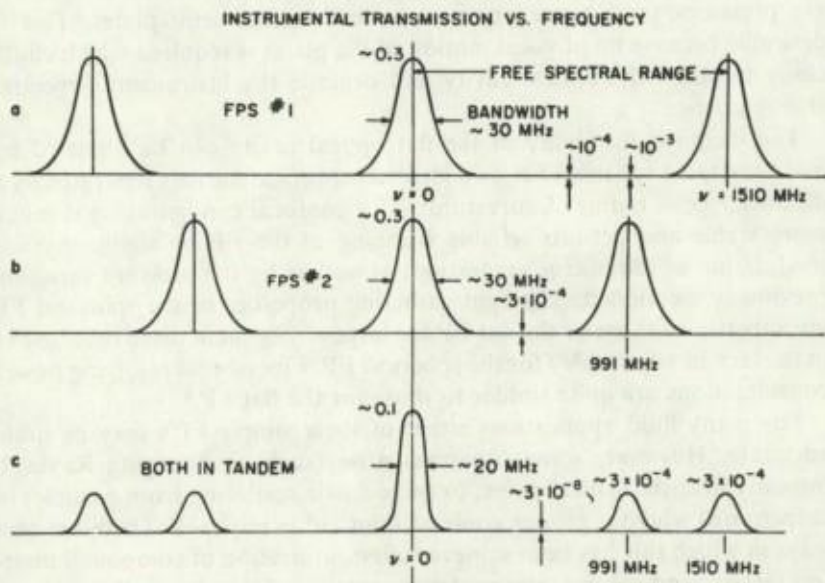


Fig. 6. Transmission patterns of single (*a* and *b*) Fabry-Perots individually and (*c*) in a tandem arrangement. After D. S. Cannell, Thesis, Massachusetts Institute of Technology, Cambridge, Mass., 1970, unpublished.

resolving power improved as well. The instrumental response function is considerably more complicated and construction more difficult than for a single FP, but the improvements permit experiments not otherwise possible. Cannell's application of his tandem FP to the study of the critical point in xenon is discussed in Section IV below.

A third and perhaps simpler method of enhancing contrast is that of the multiply passed FP, utilized recently by Sandercock.²⁶ By sending the optical signal through the same flat FP several times he has demonstrated

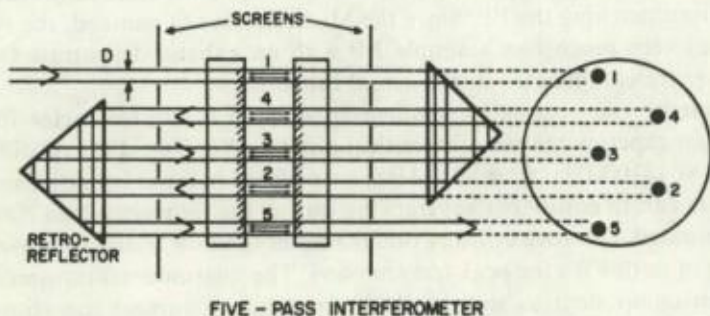


Fig. 7. Design of multiple-pass flat FP (Sandercock²⁶).

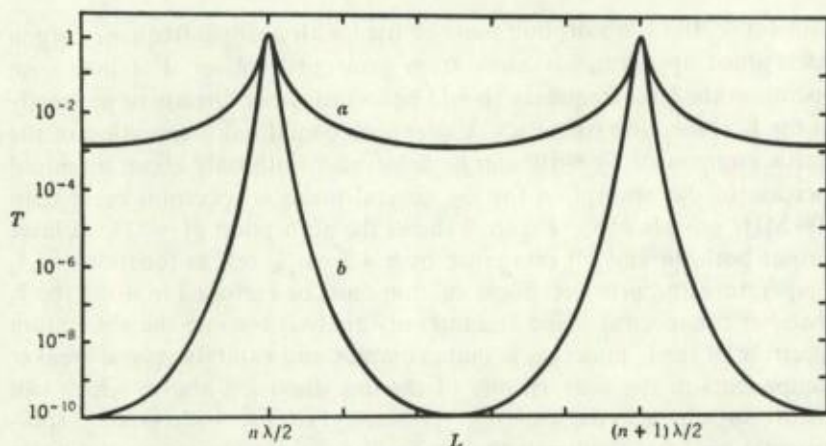


Fig. 8. Theoretical transmission curves for the above instrument used in (a) single-pass and (b) five-pass arrangements, assuming single-pass finesse of 40 (Sandercock²⁸). Note log scale.

enormous contrast enhancements together with an increased finesse. Since only one pair of plates is involved, scanning, alignment and stability requirements are essentially those of a single FP. The requirement that each beam pass must use a different area of the plates provides some restrictions on light gathering power and overall instrumental transmission. The combination of various design requirements, including mirror loss, reflectivity, flatness, and overall transmission, has been shown to suggest a five-pass instrument as a practical optimum. Such an arrangement together with its theoretical transmission curve (compared to a single FP) are sketched in Figs. 7 and 8. These are for flatness of $\lambda/100$, 0.2% mirror absorption, and $0.86 = R$. The overall finesse is 40 and the contrast greater than 10^9 . This kind of instrument should open several areas to experimental perusal.

2. Resonant Reabsorption

The *effective* contrast of a detection system can also be enhanced by reducing the intensity of the elastic component relative to that of the inelastic components prior to detection of the spectrum. In principle a very narrow absorption line, were it to coincide with the exciting laser frequency, would accomplish this. Unfortunately such coincidences are rare. However, nature has been kind in providing a strong and very narrow absorption line in the rotation-vibration spectrum of I_2 vapor which falls within the gain curve for the 5145 Å line of the argon ion laser. Because it is so narrow (<300 MHz) and slightly shifted in frequency from the argon

gain curve, the I_2 absorption must be used with a single-frequency argon laser tuned appropriately away from gain curve center. For long-term operation the laser frequency should be locked either directly or indirectly to the I_2 absorption frequency. Under such conditions attenuation of the elastic component by $\sim 10^8$ can be achieved,²⁰ with only about a tenfold increase in the absorption for the general inelastic spectrum more than 300 MHz outside of ν_{I_2} . Figure 9 shows the absorption of ~ 5145 Å laser output both on and off resonance by a 4.5 cm I_2 cell as functions of I_2 temperature and pressure. Some caution must be exercised in using the I_2 absorber for spectral shape and intensity analysis because the absorption spectrum of the I_2 molecule is quite complex and exhibits several weaker components in the near vicinity of the line discussed above, which can distort or obscure the *inelastic* spectrum. For use with grating spectrometer we have compared the observed quasicontinuous second-order Raman spectra in $SrTiO_3$ with and without the I_2 absorption cell (see Fig. 10). Similar calibration of the I_2 absorption in the lower-frequency range has been obtained by doing the same comparison on the Rayleigh wing spectrum of CS_2 . In both cases the size, shape, and position of the "holes" eaten out of the spectrum reveal the effects of the resonant I_2 absorber. For studying weak inelastic spectra, fluorescence from the I_2 cell, induced by the absorption of the elastic component, can produce spurious lines in the spectral region of interest. So the improvement in *effective* contrast achieved with an I_2 cell is accompanied by a considerable complication in the overall instrumental response function.

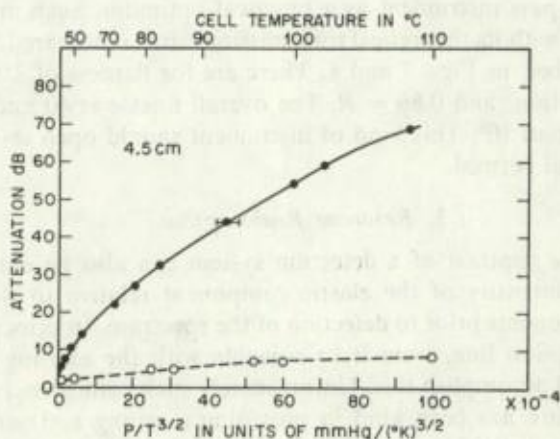


Fig. 9. Temperature and pressure dependence of resonant I_2 absorption for suitably tuned single-mode Ar laser at 5145 Å, on (solid) and off (dashed) resonance. After Devlin et al.²⁰

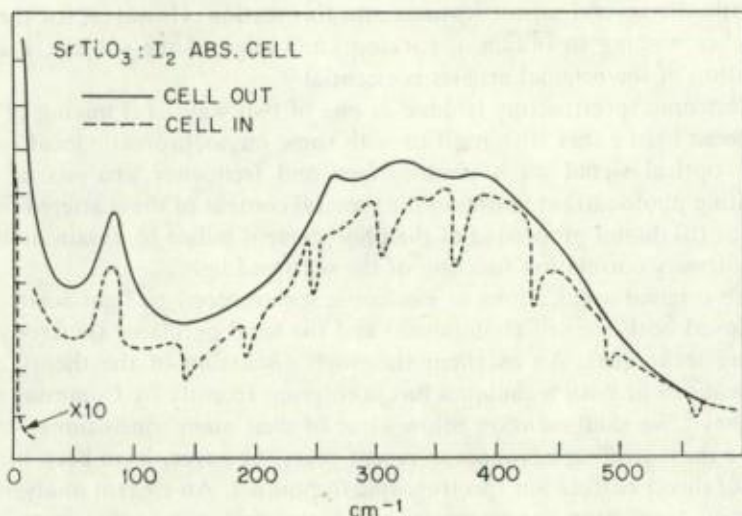


Fig. 10. Effect of subsidiary I_2 absorptions on inelastically scattered light shifted $0 \rightarrow 500 \text{ cm}^{-1}$ from 5145 Å laser line. Illustrated by room-temperature second-order Raman spectrum of SrTiO_3 .

Finally it should be mentioned that other absorbers exist for other laser lines (CH_4 for 3.39μ HeNe; SF_6 for 10.6μ CO_2 , etc.). Naturally the combination of the I_2 absorber and one of the compound interferometers discussed earlier would be an obvious extension.

C. Low-Frequency Shifts

For the highest-resolution experiments of interest in fluids, optical spectrometers or interferometers are inadequate, and direct electronic processing of the optical signal must be employed. Basically these involve measuring the intensity correlation function (or its Fourier transform) of the scattered light via a nonlinear detector (most often a photomultiplier tube). Frequencies ranging from $<1 \text{ Hz}$ to $>1 \text{ MHz}$ can thus be measured, corresponding to a resolving power between 10^9 and 10^{15} for visible laser sources. Whereas the optical spectrometers and interferometers discussed above were widely employed before the advent of laser sources, the electronic spectrometers considered here require for successful operation source brightnesses obtainable only with lasers. Thus the literature on such spectrometers is relatively sparse and many of the concepts are unfamiliar to students of more traditional spectroscopy.

Recently several very useful papers have appeared which explicate the theory and practice of electronic spectroscopy and from which we shall

incorporate several salient features into this section. However, for the researcher wishing to obtain a working knowledge of the subject, a consultation of the original articles is essential.

Electronic spectroscopy is done in one of two ways: (a) mixing of the scattered light either with itself or with some monochromatic local oscillator optical signal on a photosurface and frequency analysis of the resulting photocurrent to obtain the spectral content of the scattered field, and/or (b) digital processing of the photocurrent pulses to obtain directly the intensity correlation function of the scattered light.

The original applications of electronic spectroscopy to light scattering employed both the self (homodyne) and the local oscillator (heterodyne) mixing techniques. An excellent thorough discussion of the theory and applications of both techniques has been given recently by Cummins and Swinney.³ We shall mention below some of their main conclusions.

The most striking advances in recent years, however, have been in the area of direct correlation spectroscopic techniques. An elegant analysis of intensity correlation spectrometers and their performance in comparison with light beating spectrometers has been given by Degiorgio and Lastovka.¹⁹ Additional considerations on the theory of correlators by Jakeman and Pike²¹ have also appeared. Although much of this analysis is for experts, we shall list some of these basic conclusions here as well.

Let us first consider the basic expression for the scattered intensity in a form amenable to electronic spectroscopy.³ The utility of electronic spectroscopy rests upon the quantitative relationship between the spectrum or statistical properties of the scattered optical field and the spectrum or count distribution of the photoelectric current. This relationship is simple for an optical field which is a Gaussian random process and not so generally otherwise. Fortunately even when the incident light is coherent, the light scattered from thermal fluctuations is Gaussian, so we can proceed on that assumption.

We note that the instantaneous intensity at the photocathode $I(t)$ is just $E^*(t)E(t)$. Assuming the field to be spatially coherent over the illuminated photocathode area the photocurrent will be $i(t) = \sigma E^*(t)E(t)$ or

$$\langle i(t) \rangle = \sigma \langle i \rangle = \sigma \langle E^*(t)E(t) \rangle \quad (89)$$

It is also important to define the correlation function between the photocurrent at t and at $t + \Delta$

$$\begin{aligned} \langle i^{(2)}(t, t + \Delta t) \rangle &= \sigma^2 \langle E^*(t)E(t)E^*(t + \Delta t)E(t + \Delta t) \rangle \\ &= \sigma^2 \langle I \rangle^2 g^{(2)}(\Delta t) \end{aligned} \quad (90)$$

$$g^{(2)}(\Delta t) \equiv \frac{\langle E^*(t)E(t)E^*(t + \Delta t)E(t + \Delta t) \rangle}{\langle E^*E \rangle^2} \quad (91)$$

The scattered spectrum is just the Fourier transform of

$$\langle E^*(t)E(t + \Delta\tau) \rangle = \langle I \rangle g^{(1)}(\Delta\tau) \quad (92)$$

Now for Gaussian fields the correlation functions $g^{(1)}$ and $g^{(2)}$ are simply related

$$g^{(2)}(\tau) = 1 + |g^{(1)}(\tau)|^2 \quad (93)$$

The photocurrent correlation including the self-term can then be expressed³:

$$\begin{aligned} C_i(\tau) &= e^{i\omega_0\tau} \delta(\tau) + \langle i^2 \rangle g^{(2)}(\tau) \\ &= e_{(i)} \delta(\tau) + \langle i^2 \rangle (1 + |g^{(1)}(\tau)|^2) \end{aligned} \quad (94)$$

The photocurrent spectrum $P_i(\omega)$ is just the Fourier transform of $C_i(\tau)$, and the homodyne and heterodyne situations may be distinguished as follows. For homodyning the total field consists of the scattered field alone (no local oscillator). Its correlation function is given by $g^{(1)}(\Delta\tau)$ in (92), which is simply derivable from the photocurrent correlation function (94). Traditionally it has been the power spectrum of the photocurrent, that is, the Fourier transform of $C_i(\tau)$, which has been measured. This measurement involves feeding the photocurrent into a spectrum analyzer which then sweeps a narrow frequency window across the frequency range, viewing sequentially the various portions of $P_i(\omega)$. In the last few years, however, the direct measurement of $C_i(\tau)$ itself through the use of digital correlators has become increasingly popular. Although there are many subtleties in comparing the two approaches, it is generally true that, all other things being equal, the rate of data accumulation is faster in the correlation approach by a factor of order $(\Delta\omega\tau_c)^{-1}$ where τ_c is the correlation time and $\Delta\omega$ the frequency window width for the spectrum analyzer. Some other aspects of direct correlation will be discussed below.

For the heterodyne method the field appearing in (92) consists of the sum of the scattered field $E_s(t)$ and a coherent local oscillator field $E_0 e^{i\omega_0 t}$ usually derived from the incident laser beam itself. The current correlation $C_i(\tau)$ is therefore a more complicated expression than for the homodyne case ($E_0 = 0$). In the limit where $E_0 \gg E_s$ we have³

$$\begin{aligned} C_i(\tau) &= e^2 \sigma I_0 \delta(\tau) + e^2 \sigma^2 I_0^2 + e^2 \sigma^2 I_0 \langle I_s \rangle \\ &\quad \times \{ e^{i\omega_0\tau} \langle E_s^*(t)E_s(t + \tau) \rangle + e^{-i\omega_0\tau} \langle E_s(t)E_s^*(t + \tau) \rangle \} \\ &= e i_0 \delta(\tau) + i_0^2 + i_0 \langle i_s \rangle \{ e^{i\omega_0\tau} g^{(1)}(\tau) + e^{-i\omega_0\tau} g^{*(1)}(\tau) \} \end{aligned} \quad (95)$$

Notice in this case that the relation between $C_i(\tau)$ and $g^{(1)}(\tau)$ does not proceed through $g^{(2)}(\tau)$ and thus holds even for non-Gaussian statistics. The $P_i(\omega)$ resulting from (95) is an exact replica of the optical spectrum centered at a frequency $(\omega_1 - \omega_0)$, which is zero if the incident laser is

used as the local oscillator. These latter attractive features of heterodyning are somewhat offset in practice by the requirement that the fields E_0 and E_s must be parallel and coincident at the photosurface to prevent drastic decrease in the mixing efficiency. This presents severe alignment difficulties not present in the homodyne scheme.

As with homodyning the photocurrent may be either Fourier analyzed for $P_i(\omega)$ or processed directly to obtain the correlation function $C_i(\tau)$. Again the rate of data accumulation is much faster with the correlator than with the spectrum analyzer scheme. Recently multichannel spectrum analyzers have become available which permit simultaneous observation of all frequency intervals, rather than requiring sequential sampling of a swept, "single-channel" analyzer. Such devices recover the factor $(\Delta\omega\tau_c)^{-1}$ in the data accumulation rate mentioned above. The operation of spectrum analyzers is discussed more fully by Cummins and Swinney³ and by Benedek.²¹⁷

Let us now turn briefly to a description of digital correlators so as to appreciate the recent advance represented by the so-called "clipped" correlator.¹⁹ The photocurrent in a photomultiplier is generated by the arrival of each photon at the photocathode causing the emission of a photoelectron, which is then accelerated through the dynode chain where it produces additional (roughly simultaneous) electrons, all of which, perhaps a million in all, arrive in a bunch at the tube anode, giving rise to a "pulse" of photocurrent. If the photon arrival rate (light intensity) is not too great each photon will give rise to a photoelectron pulse arriving at the anode at a precisely delayed time after the photon struck the photocathode. Ignoring the statistical complications of (a) the initial photoemission process and (b) the photoelectron amplification process, one would expect the arrival rate and distribution of photocurrent pulses at the anode to replicate accurately those of the photon arrivals at the cathode. The photocurrent, conveniently viewed as an analogue signal for light beating purposes, is in reality a digital signal, quite amenable to digital correlation processing. Degiorgio and Lastovka¹⁹ have described the operation of an ideal digital correlator as follows: divide the total measurement time T into M_0 equal intervals of duration Δt . Define η_j as the number of counts (photocurrent pulses assumed identical in size and shape) recorded between $j\Delta t$ and $j\Delta t + \delta t$ (with $\delta t < \Delta t$). The photocurrent autocorrelation function (at $l\Delta t$) is then

$$R_l = \frac{1}{M_0} \sum_{j=1}^{M_0} \eta_j \eta_{j+l} \quad (96)$$

For a Gaussian field this has the form

$$R_l = \langle n \rangle^2 (1 + e^{-l\Delta t/\tau_c}) \quad (97)$$

where τ_c is the correlation time. Naturally to achieve good representation of R_l in a reasonable T , one arranges for $\Delta t \ll \tau_c \ll T/5$. The time and circuiting required for formation of the full product pairs in (96) would make a many-channel (~ 100) full correlator quite complicated, expensive, and slower than the simple clipped correlator described below.

Present-day electronic components can perform simple logical operations as quickly as $\sim 10^{-9}$ sec and permit in principle comparably small values of Δt and measurements of τ_c as small as 3×10^{-8} sec. This corresponds to frequencies as high as 10 MHz and clearly overlaps the low-frequency range of Fabry-Perot spectrometers. Measurement of longer τ_c , or lower frequencies, down to < 1 Hz are easy with correlators. However, a machine that will accurately perform the full correlation operation indicated in (96) requires many more logical operations and is thus slower than the simpler, nearly as accurate, device we now describe.

It has recently been shown that essentially the same information and accuracy are available from a much simpler device called a clipped correlator.¹⁹ The basic simplifying operation is to introduce a positive integer clipping level, k , for the number of counts collected in interval δt . Thus we can define a clipped count $n^{(k)}$ as

$$\begin{aligned} n^{(k)} &= 1 & n > k \\ &= 0 & n \leq k \end{aligned}$$

The clipped correlator then operates in the following truncated manner¹⁹: "at $t = 0$ the correlator begins sampling photocurrent with gates of duration $\delta t \leq \Delta t$. The first gate in which the collected number of counts exceeds k triggers an N -channel scaler which records sequentially the number of counts during each of the N gates immediately following the trigger gate. After N gates the trigger is again started on the next $n > k$ count at the first channel. The process is then repeated a total of M_0 times with the number of counts in each channel added to the total in that channel from previous triggers. After M_0 triggers the l th channel contains the sum of all counts registered in all the M_0 gates occurring $l \Delta t$ after the trigger." The clipped correlation function thus formed is

$$R_l^k = \frac{1}{M_0} \sum_{m=1}^{M_0} n^{(k)}(t_m) n(t_m + l \Delta t) \quad (98)$$

where n is the full count and n^k the clipped count. The operations involved in forming the sum, then, multiplication by only 1 or 0 and addition of the results to the contents of a register, are much less complicated than in the

full correlator. Jakeman and Pike²¹ have computed the clipped correlation function for a Gaussian exponential field and found

$$R_k(t) = \langle n \rangle \left(\frac{\langle n \rangle}{1 + \langle n \rangle} \right)^{k+1} \left[1 + \left(\frac{1+k}{1+\langle n \rangle} \right) e^{-t/\tau_c} \right] \quad (99)$$

The similarity in form to that of (96) is striking and suggests how τ_c may be extracted from either form with equal ease.

A detailed analysis and comparison of the statistical errors inherent with both the ideal and clipped digital correlators, as well as the homodyne spectrum analyzer, have been given by Degiorgio and Lastovka.¹⁹ One of their comparisons is summarized in Fig. 11, where the fractional rms statistical errors involved in determining τ_c are plotted for the ideal and the clipped correlators and for single-channel and 100-channel spectrum analyzers as a function of the counting rate (expressed in number of counts per correlation time = $\eta\tau_c > 1$). The clipped correlator is better than the spectrum analyzer and only slightly worse than the full correlator.

Finally, one crucial concept which must be mentioned is that of the "coherence area." All the above discussion has presumed that the optical field is spatially coherent over the surface area of the photodetector. As long as this is true the signal to shot noise ratio increases linearly with detector area. For a signal derived from an incoherent scattering process, the scattered field source is spatially incoherent. The result is that the scattered field is only coherent over a limited area $A_c = 2\lambda^2/\Omega$, where λ is the optical wavelength and Ω is the solid angle subtended by the source

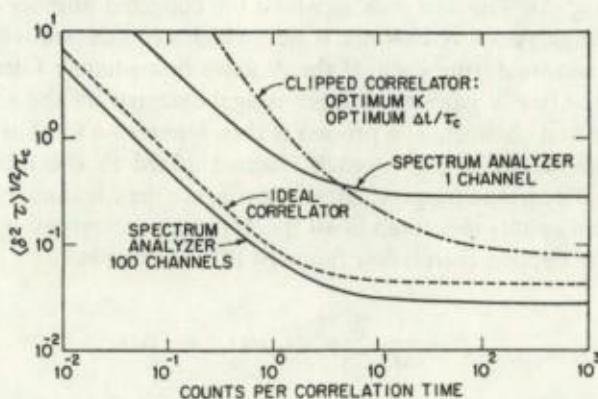


Fig. 11. RMS fractional errors vs. counting rate per correlation time for clipped and ideal correlators and for single- and 100-channel spectrum analyzers. After Degiorgio and Lastovka.¹⁹

at the detector. For a detector area $A = \alpha A_c$, the α elements act independently so far as mixing is concerned, with the result that increase in A beyond A_c causes shot noise to increase as rapidly as signal with no change in the ratio. Under certain conditions, in fact, the error in the $C_i(\tau)$ measurement actually *increases* with larger α whereas the error in $P_i(\omega)$ is independent of α .¹⁹

IV. EXPERIMENTAL RESULTS

So active is the research on light scattering in fluids that several new advances have been made which have not been discussed in even quite recent review articles. In this section we shall emphasize these, referring to earlier work only where necessary to provide a suitable context in which to view the more recent results. As we have seen in the earlier theoretical sections, the richness of spectral information obtainable from a fluid system increases as the system becomes more complex. In Fig. 12 we have tried to represent this situation schematically for fluid systems ranging in complexity from a simple monatomic gas to a mixture of polyatomic liquids. In the latter case the spectra are so complex, especially when one considers the possibility of interaction effects among the various degrees of freedom, that theoretical treatments are still on the level of phenomenology. Nevertheless it is hoped that by comparing in sufficient detail the spectra of successively more complex fluid systems we may be able to "boot strap" our way to a more fundamental understanding of the liquid state.

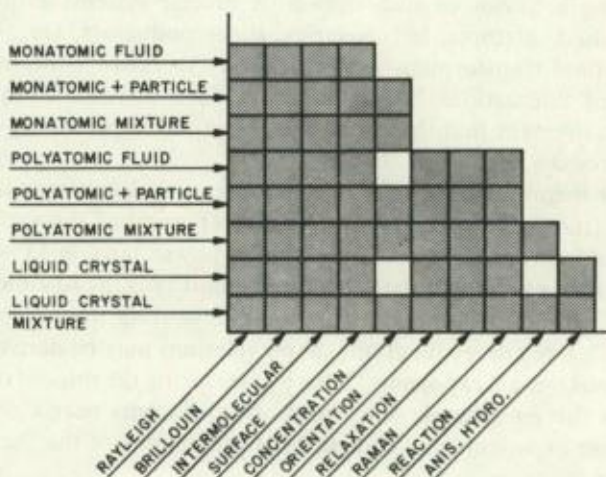


Fig. 12. Schematic illustration of contributions to spectra (shaded blocks) from various processes (horizontal axis) for fluids of increasing complexity (vertical axis).

As can be seen from Fig. 12 all fluid systems support density fluctuations, which in general give rise to Brillouin scattering from the thermal sound waves (adiabatic density fluctuations) and to Rayleigh scattering from isobaric entropy fluctuations. The Brillouin spectrum measures the velocity and attenuation of high-frequency sound, whereas the Rayleigh spectrum, centered at zero frequency shift, has a width proportional to the thermal diffusivity, $\kappa/\rho C_p$. In superfluid helium the isobaric entropy fluctuations are propagating rather than purely dissipative, and their spectra provide a measure of the velocity and attenuation of second sound.

A. Liquid Gas Critical Point

Much of the recent work on the Rayleigh-Brillouin spectra of pure fluids has been devoted to the vicinity of the liquid-gas critical point (C.P.). It has long been realized that near C.P. the Rayleigh peak will narrow in width and increase in intensity (critical opalescence), due mainly to the divergence in the specific heat and density correlation length. The precise manner of this behavior has been of considerable interest in testing the so-called "scaling laws" proposed for second-order phase transitions. According to the scaling hypothesis sufficiently near C.P. there is only one fundamental length whose behavior depends only on general symmetry properties of the phase transition and not on the details of interparticle interactions. This length, called the "correlation length," ξ , and its behavior in terms of $|X - X_c|$ (where X is some appropriate thermodynamic variable, like temperature or density) provide a convenient means of unifying the behavior of such outwardly diverse systems as pure fluids, magnets, fluid mixtures, ferroelectrics, superconductors, etc., when they undergo phase transformation. It also serves to connect the near-critical behavior of fluctuations having different wave vectors, k , through the scaling requirement that the correlation functions depend only on dimensionless product $k\xi$.

Another theoretical expectation regarding critical behavior is that very near C.P. (the "asymptotic region") the divergences of the various correlation functions are describable by simple power laws in $|X - X_c|$. The value of the power appropriate to a given quantity is the so-called "critical exponent." Based on general assumptions regarding the homogeneity of the system's free energy function, simple relations may be derived between these various critical exponents. Thus by measuring the rates of divergences of various thermodynamic and transport coefficients near C.P. one may obtain these exponents and thus test the predictions of the "scaling law" hypothesis.

Very recently a rather long-standing controversy regarding the apparently different critical behavior of SF₆ from that of simpler fluids like Xe and

CO₂ has been resolved. In the hydrodynamic regime and for measurements along the critical isochore, the Rayleigh line width measures the ratio $\kappa/\rho C_p$ and thus a combination of critical exponents $(\gamma - \nu) \equiv \varphi$ since $C_p \propto \varepsilon^{-\gamma}$ and $\kappa \propto \varepsilon^{-\nu}$ (where $\varepsilon = |T - T_c|/T_c$). Additional information is provided by the frequency integral of the scattered intensity $I(k) = [AK_T \sin^2 \alpha / (1 + k^2 \xi^2)]$. The angular dependence of $I(k)$ measures ξ , and the angular integrated intensity I measures K_T , the isothermal compressibility. These are described by $\xi = \xi_0 \varepsilon^{-\nu}$ and $K_T = K_0 \varepsilon^{-\gamma}$, respectively.

Several years ago Saxman and Benedek²¹⁷ obtained in SF₆ a Rayleigh line width which vanished as $\Gamma_R \propto \varepsilon^{1.26}$; that is, $\varphi = 1.26$. Later measurements for the simple fluids CO₂ and Xe revealed apparent φ 's of 0.73^{81,82} and 0.751,⁵⁸ respectively. This striking disagreement appeared to contradict the expected universal behavior of critical exponents. Thus many other experiments were done on these three fluids to isolate the source of the discrepancy. Direct measurements of γ showed values for all three fluids of $\sim 1.22 \pm 0.02$; for ν the results were all consistent with $\sim 0.63 \pm 0.07$. The original value for φ for SF₆ became increasingly suspect. Recently three independent remeasurements of the Rayleigh line width in SF₆ were made^{40,51,63} which yielded values for φ quite close to those for CO₂ and Xe. It now appears that the original SF₆ data were in error, although, because they represent *too narrow* a Rayleigh width for a given value of ε , the exact experimental source of that error remains unknown.

Paradoxically the existence of the error itself has probably had a positive effect on our understanding of critical phenomena, since the anomalous φ for SF₆ forced more careful consideration of critical phenomena and the subtleties of data analysis than might have otherwise occurred. One result of this effort has been the realization that the Rayleigh width $Dk^2 (D = \kappa/\rho C_p)$ receives a non-negligible contribution from the non-singular or background thermal conductivity κ_B . Thus $k^2 \kappa_B / \rho C_p$ must be subtracted from the measured line width in order to obtain $k^2 \kappa_S / \rho C_p$, the true singular part. When done for the above experiments, values of 0.61 ± 0.04 ,^{51,63} 0.62 ,^{81,82} and 0.64 ± 0.04 ⁵⁸ for φ are obtained for SF₆, CO₂, and Xe, respectively. Not only are these exponents all essentially the same, but furthermore they are in remarkably close agreement with a recent theoretical prediction by Kawasaki⁵⁰⁸ that the singular part should be given by $k^2 k T / 6\pi\eta\xi$. That is, the singular contribution to the Rayleigh width is just that corresponding to the mass diffusion of a fluid sphere with radius $r = \xi =$ the correlation length. Thus φ should be equal to $-\nu$ according to Kawasaki, and the above corrected φ values are quite consistent with the previously quoted $-\nu = 0.63 \pm 0.07$.

B. Critical Mixing

The behavior of Brillouin spectra near C.P. has also been studied in detail for some pure fluids. However, before discussing these results we shall briefly consider the critical opalescence studies made in two-component fluid mixtures. As seen in Section II the spectrum of a solution or a fluid mixture exhibits an additional central frequency component due to fluctuations in the relative concentrations of the two species. Away from critical points where correlation lengths are small, this component is Lorentzian in shape with a width $\Gamma_c = Dk^2$, where D is (a) the solute mass diffusion coefficient for a dilute solution or (b) $\alpha(\partial\mu/\partial c)$ for a concentrated mixture, where α is a transport coefficient, c is the concentration, and μ is the chemical potential. For binary mixtures near the critical mixing point, concentration fluctuations are correlated over a sufficient range that $k\xi$ may no longer be $\ll 1$. Then the line width is $\Gamma_c = D_c k^2(1 + k^2\xi^2)$. Chu has done extensive studies of line width as well as of angular and total intensity for several binary mixtures and has found that the critical exponents agree well with their counterparts in the liquid-gas critical transition.^{2,305,306,354} Swift⁵⁰⁷ has predicted that k^2D should behave for binary mixtures with the same exponent that the Rayleigh line width does in a pure fluid near C.P.; that is, $D \propto \epsilon^{\nu-\nu}$. The line widths measured for a variety of binary mixtures² yield exponents of $\sim 0.66 \pm 0.05$ on the average, in good agreement with Swift's modest proposal. Of course the critical behavior of ξ may also be measured by angular dependence of the scattered intensity. The results thus obtained are in good agreement with the line width measurements.

C. Brillouin Scattering

1. Simple Fluids

A few Brillouin experiments in simple monatomic liquids away from C.P. have been performed with the aim of possibly observing some of the departures from Navier-Stokes theory predicted by the generalized hydrodynamic analysis of Section II. The studies of Fleury and Boon¹⁰⁷ on argon and neon initially suggested the existence of a small negative dispersion in the hypersonic velocity for sound frequencies in the 1-3 GHz range. Although close in size to the experimental error, the observed velocity difference from low-frequency (< 1 MHz) values subsequently measured in neon⁵⁰⁶ supports the need for more accurate Brillouin and ultrasonic measurements in simple liquids. A simple corresponding-states argument (see Section II.3) suggests that if the $\sim 1\%$ negative dispersion in neon were due to quantum effects, rather than to finite frequency effects, then

normal liquid helium should exhibit an even larger dispersion.¹⁵⁹ Very recently Pike et al.⁷⁸ have observed the Brillouin spectrum from normal liquid helium, and find no dispersion greater than $\sim 0.5\%$ relative to velocities obtained at much lower frequencies. The origin, if not the existence, of small velocity dispersion in argon and neon then remains to be understood.

2. Complex Fluids

In fluids where there exist "internal" energy-storing degrees of freedom (such as vibrational, rotational, or configurational), relaxation processes can introduce quite large frequency dependence to both sound velocity and attenuation. These effects (discussed theoretically in Section II.3) were first studied in detail using Brillouin scattering by Chiao and Fleury^{95,108} and have since been extended by several others.^{37,38,105,137,158} Because relaxation effects are most noticeable for frequencies $\omega \approx \tau^{-1}$ and because for many relaxation processes in fluids τ^{-1} is of order 0.1–10 GHz, Brillouin scattering is an ideal way to investigate such phenomena. With increasing precision and sophistication in spectral line shape measurement and analysis has come corresponding increase in the accuracy of determining the relaxation parameters, τ and $(U_\infty^2 - U_0^2)/U_0^2$.

Perhaps the most interesting aspect to Rayleigh-Brillouin spectra arising from relaxation process is an additional central peak of width approximately τ^{-1} . Implicit in the phenomenological theory of Rytov, this feature was discussed explicitly in Mountain's hydrodynamic theory of 1966^{11,130} and was first observed experimentally in CCl_4 at about the same time.¹¹⁵ Although since observed in several relaxing fluids,¹²¹ this relaxation mode has not proved as helpful in providing a direct measure of relaxational parameters as was initially hoped. This is primarily due to its rather broad shape, difficult to extract precisely in the presence of the stronger Rayleigh and Brillouin components, at least with the instrumental resolution applied thus far.

It has been realized recently by a number of people that the relaxational peak and the form derived by Mountain are special-case manifestations of a rather general phenomenon not restricted to relaxing liquids, but present in magnets, ferroelectrics, superconductors, etc., as well. The existence of an additional central peak in the scattering response function $S(k, \omega)$ of a system can generally be ascribed to a frequency-dependent damping of the mode of interest. The shape and strength of this central peak are determined by the precise shape and strength of this frequency dependence. One can write the susceptibility $\chi(\omega) = [\omega_0^2 - \omega^2 + i\Gamma(\omega)\omega]^{-1}$. Then

$$S(\omega) = -\frac{n+1}{\pi} \text{Im } \chi = \frac{(n+1)\Gamma'\omega}{\pi([\omega_0^2 - \omega^2 - \omega\Gamma']^2 + \omega^2\Gamma'^2)}$$

where $\Gamma = \Gamma' + i\Gamma''$. For relaxation behavior $\Gamma(\omega) = \Gamma_0/(1 + i\omega\tau)$. The resulting $S(\omega)$ takes on different forms depending on the values of $\omega\tau$ and $\Gamma_0\tau^{-1}$. The limit most appropriate to Brillouin scattering in relaxing fluids has $\tau^{-1} < U_\infty q$ and $(U_\infty^2 - U_0^2)/U_\infty^2 < 1$. In this case the Brillouin peaks contribute a fraction $\sim [1 - (U_0^2/U_\infty^2)]$ of their strength to the relaxation peak and it will be approximately Lorentzian in shape, of width τ^{-1} . [($n + 1$) is the thermal population factor.]

The relaxation peak would appear much more striking in the limits $(U_\infty^2 - U_0^2)/U_0^2 > 1$; $\tau^{-1} < U_\infty q$. Then the central peak will carry the major fraction of the strength, $[1 - (U_0^2/U_\infty^2)]$, and will have a width $\omega_0^2/\Gamma_0 = U_0^2 q^2/\Gamma_0$.

Passage from one region to another is usually not possible for Brillouin scattering in a relaxing liquid, because τ^{-1} and Γ_0 are not usually sufficiently strong functions of temperature, pressure, etc. An exception to this occurs in the vicinity of phase transitions. In particular, at the C.P. in a simple monatomic fluid, the fluid correlation volume elements themselves play the role of the internal degrees of freedom, giving rise theoretically to two distinct relaxation times τ_R and τ_B . $\tau_R^{-1} = (\kappa/\rho C_p)\xi^{-2}$ corresponds to the inverse time required for thermal diffusion to occur over a distance of one correlation length. $\tau_B = \xi/U$ corresponds to the time required for a sound wave to cross the correlation length. Both these relaxations vary greatly as $T \rightarrow T_c$. Cannell⁹⁰ has probed the region $\omega\tau_B \approx 1$ in his Brillouin studies near xenon's C.P. and has observed the central relaxational peak quite near C.P. The sound velocity dispersion accompanying the τ_B relaxation has been well documented in this and earlier ultrasonic studies. Other Brillouin studies near C.P. have been carried out by Gammon et al.¹¹¹ and by Ford et al.¹⁰⁹ in CO_2 , and by Mohr et al.¹²⁷ in SF_6 .

All these experiments obtained similar results and had to overcome similar difficulties, mainly arising from the diverging Rayleigh intensity as the C.P. is approached. In all experiments effects of relaxation phenomena on the hypersonic critical behavior were evident, and thus complicated considerably comparisons with low-frequency sound experiments. The correlation-length-related relaxation processes in Xe already mentioned are present in CO_2 and SF_6 as well. However, these polyatomic fluids exhibit in addition vibrational and rotational relaxations. The result is that although "exponents" describing both the Brillouin splitting and line width dependences on $(T - T_c)$ can be extracted from these data, their values are generally frequency dependent, are not the same for the different fluids, and thus are not directly relevant to tests of theoretical scaling predictions. Xenon presents by far the simplest case, and Cannell⁹⁰ has used the full spectral shapes to determine not only the hypersound velocity and attenuation, but several other parameters as well, including C_p/C_v , τ_B ,

$\kappa/\rho C_v$, and U_0 . His discussion clearly demonstrates the much greater difficulty in understanding Brillouin spectra near C.P. than the Rayleigh spectrum. There is room for much work on this problem.

Even more complicated to understand microscopically is the Brillouin spectrum near the solution critical point of a binary liquid mixture. Arefev⁸⁶ has observed the Brillouin spectrum of triethylamine (44.6% wt) and water near its critical mixing temperature, 17.9°C. A strongly temperature-dependent velocity dispersion was observed, but no definitive interpretation could be made. Similar results were obtained for the nitrobenzene-*n*-hexane mixture studied by Chen and Polonsky.³⁶⁰ In contrast to the behavior in the pure fluid C.P. experiments, their results show the Brillouin line width continuing to diverge for $T \rightarrow T_c$ rather than leveling off for $\Delta T < \sim 0.7^\circ\text{C}$, as observed in pure fluids.^{109,111,127} Again the spectra receive contributions from relaxation processes associated with the correlation range of the concentration fluctuations, as well as from the noncritical molecular relaxations due to the polyatomic nature of the liquid constituents. Again, much more detailed experimental work on Brillouin spectra of critical mixtures is required to put understanding on a quantitative basis. It seems appropriate to point out here that the Rayleigh-Brillouin spectrum of a critical mixture should exhibit an additional central "relaxation" component. No such component has yet been reported, and a search for it might be of interest.

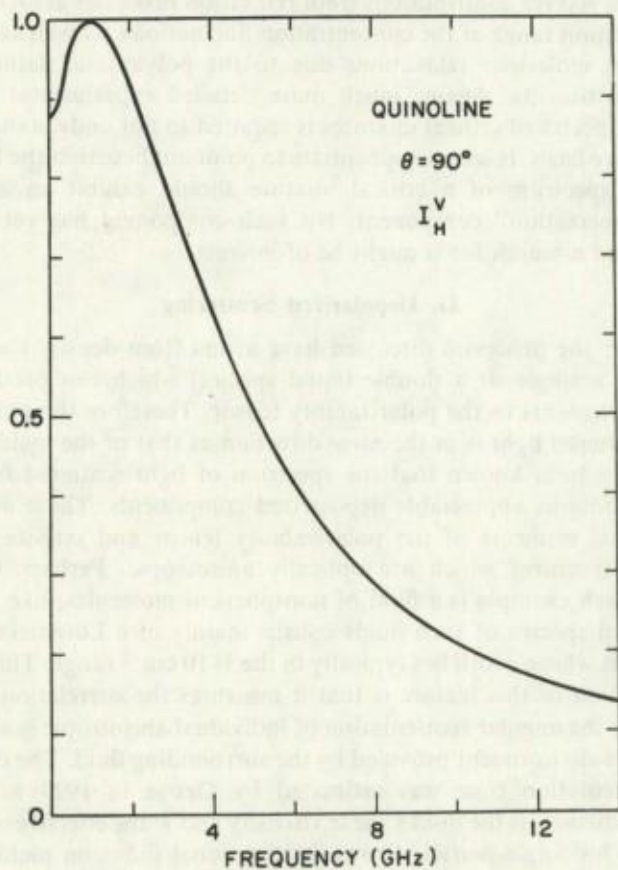
D. Depolarized Scattering

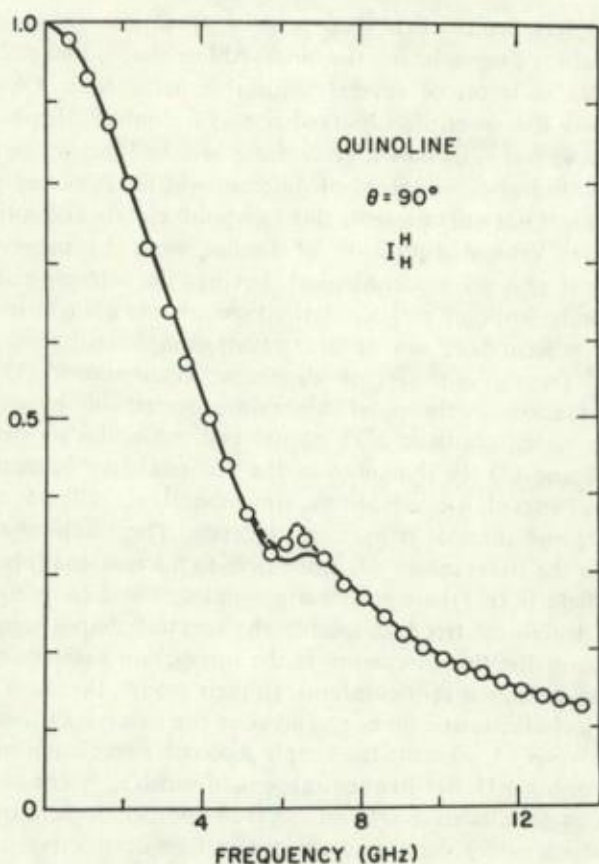
Thus far the processes discussed have arisen from density fluctuations (of either a single or a double liquid species) which can produce only diagonal elements to the polarizability tensor. Therefore the polarization of the scattered light is in the *same* direction as that of the incident light. It has long been known that the spectrum of light scattered from fluid systems contains appreciable depolarized components. These arise from off-diagonal elements of the polarizability tensor and require dynamic physical structures which are optically anisotropic. Perhaps the most familiar such example is a fluid of nonspherical molecules, like CS_2 . The depolarized spectra of such fluids consist mainly of a Lorentzian central component whose width lies typically in the 1–10 cm^{-1} range. The simplest interpretation of this feature is that it measures the correlation function describing the angular reorientation of individual anisotropic molecules in the viscous environment provided by the surrounding fluid. The characteristic reorientation time was estimated by Debye in 1929 to be $\tau = \eta_s V/kT$, where η_s is the fluid's shear viscosity and V the effective molecular volume. This single-particle Brownian rotational diffusion picture dominated spectroscopists' interpretation of depolarized spectra until a very

few years ago, when two more sophisticated concepts were recognized. For identification purposes we shall call these (1) "dynamic shear waves," and (2) "intermolecular light scattering," respectively.

1. *Dynamic Shear Waves*

Whatever ultimate physical interpretation is ascribed to it, the former process definitely illustrates the inadequacy of the Debye model cited above. In 1967 high-resolution depolarized spectra of nitrobenzene and quinoline revealed an anomalous doublet structure^{254,256} (see Fig. 13). Further experiments by Stoicheff and his co-workers^{259,260} revealed similar structure in a wide variety of anisotropic molecular liquids. Since these fluids are "viscoelastic" media, one is tempted to interpret the observed structure as scattering from high-frequency shear waves in the liquid. It





(b)

Fig. 13. Depolarized spectra for quinoline observed²⁵⁹ and calculated⁴⁴ for 90° scattering in two polarization configurations. (a) Theory and experiment coincide perfectly with solid line. (b) Theories (solid and dashed curves) disagree slightly with experiment (circles).

is well known that although liquids exhibit no restoring force to the imposition of a static shear, at sufficiently high frequencies they do exhibit a nonzero (dynamic) shear modulus. Stoicheff's group has cited additional evidence for this interpretation in the k -dependence of the doublet peak splitting, which can be interpreted as $\omega_s = kV_s$, with a definite shear wave velocity V_s . This interpretation suggests that the observed doublet is associated with a collective orientation fluctuation, for which the Debye model provides the corresponding single-particle behavior.

Volterra²⁶¹ has produced a theory which quantifies these ideas and which is capable of reproducing the observed line shapes and polarization selection rules in terms of several adjustable parameters. Others have chosen to view the spectrum observed not as a doublet, displaced from zero frequency, but rather as a *central dip* arising from an *interference* between two central components of different widths. A recent paper by Chung and Yip⁴⁴ not only presents this viewpoint clearly and attractively, but contains a critical discussion of similar work by others.^{218,219,220} Their theory is also phenomenological, but has the attractive feature of unifying both the Brillouin and depolarized spectra into a single description. We will not present here any of their rather complicated spectral shape calculations. Their results rest on two basic assumptions: (1) the anisotropic fluctuations in the polarizability are describable by an effective stress tensor which contains both elastic and molecular reorientational components; and (2) the dynamics of the polarizability fluctuations are described by viscoelastic equations which simulate effects of shear, orientational, and thermal relaxation processes. The resulting equations of motion for the stress tensor ($\sigma_{\alpha\beta}$), the density fluctuations [$\rho(\mathbf{r}, t)$], and the velocity field [$v_{\alpha}(\mathbf{r}, t)$] are all linearly coupled. These couplings among the various degrees of freedom modify the spectral shapes significantly. Most significant for this discussion is the interaction between the shear stress and the molecular reorientations. In their model, the shear viscosity relaxes with a characteristic time, τ_s , and gives rise to a relaxational central peak of width $\sim \tau_s^{-1}$, whereas the simple molecular reorientation time τ_0 would cause another Debye-like central peak of width τ_0^{-1} . The coupling of these processes permits an *interference* in their contributions to the overall spectrum, which under certain conditions can be destructive, causing a central dip. This result is based on the physically reasonable assumption that $\tau_s \ll \tau_0$. For ordinary organic liquids $\tau_s \sim 10^{-12}$ sec and $\tau_0 \sim 10^{-10}$ – 10^{-11} sec. Figure 13 shows the results calculated by Chung and Yip for quinoline in two geometries. The *a* geometry does not couple to density fluctuations and the agreement with experiment is essentially perfect. The *b* geometry does couple with density fluctuations and correspondingly exhibits an interference near ω_B rather than near zero frequency. In this case agreement with experiment (open circles) is less perfect. Although there is good agreement with both polarization and scattering angle dependences of the observed spectra, this and other theories contain at least three or four adjustable parameters and must therefore be considered incomplete.

Before discussing the second type of contribution to depolarized fluid spectra, we must consider the very rapidly growing field of study represented by liquid crystals. These fluids are typically composed of very

anisotropic organic molecules which under certain conditions exhibit varying degrees of long-range orientational and translational order. These fluids may exist in several phases exhibiting increasing degrees of order: isotropic, nematic, smectic, and cholesteric. Most attention has been paid in light scattering experiments to the isotropic and the nematic phases. The former describes the normal liquid state exhibited in the anisotropic fluids discussed immediately above. The nematic phase occurs at lower temperatures and describes an arrangement where all the molecules are *orientationally* ordered (aligned), whereas their centers are translationally disordered with respect to each other. Below T_c , because of this molecular alignment the nematic liquid crystal bears many close analogies to the ferromagnetic solid. As T_c is approached from above, fluctuations in the orientational alignment Q become correlated over longer distances. Q is thus conveniently identified with the order parameter for the isotropic-nematic phase transition. ($Q = \frac{3}{2} \langle \cos^2 \theta - \frac{1}{3} \rangle$, where θ = angle between molecules' axis and local optic axis.) The spectrum of depolarized light scattered from a liquid crystal measures quite directly the fluctuations in this order parameter. Although strictly speaking this is a first-order transition, the isotropic-nematic phase transition exhibits striking pretransitional (second-order) phenomena in both its static (correlation length) and dynamic (characteristic frequencies) properties. These have been observed by measuring the integrated intensity and the frequency spectrum of the scattered light, respectively.

Among the most popular materials for optical study are *p*-methoxybenzylidene, *p*-*n*-butylaniline (MBBA), and *p*-azoxyanisole (PAA). Using a high-resolution Fabry-Perot, Litster and Stinson²⁷⁹ have measured the critical slowing down of the orientation fluctuations in MBBA above the nematic-isotropic transition. According to DeGennes' molecular field theory, the Q fluctuations in the isotropic phase decay exponentially with a characteristic time $\tau_1 = \nu/A$, where ν is proportional to the shear viscosity and $A = a(T - T_c^*)$. Stinson and Litster²⁷⁹ have observed the quasielastic Lorentzian shaped scattering whose line width $\Gamma = \tau_1^{-1}$ is shown in Fig. 14. Assuming $\nu = \nu_0 \exp(-2800^\circ/T)$ the temperature dependence of A is indeed rather linear in $T - T_c^*$. Γ was also independent of scattering angle. Another prediction of the molecular field theory borne out in these experiments pertains to the integrated intensity I_k .

$$I_k = \text{Const.} \langle Q^2(k) \rangle = \text{Const.} \frac{k_B T}{\nu a (T - T_c^*)} \frac{1}{1 + k^2 \xi^2}$$

Figure 15 shows the reciprocal intensity to be linear in $|T - T_c^*|$ for $k \simeq 10^5 \text{ cm}^{-1}$ (i.e., $k^2 \xi^2 \ll 1$).

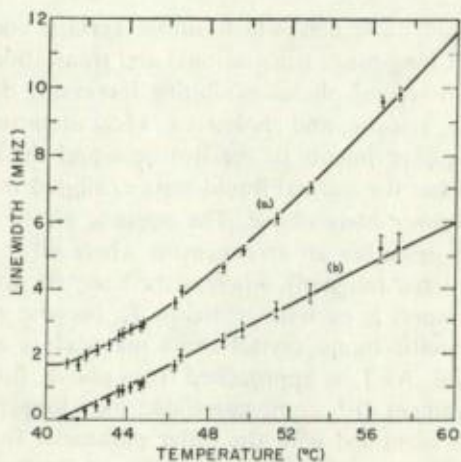


Fig. 14. Half width of central Lorentzian observed in isotropic phase of MBBA. (a) Raw data including 1.65 MHz instrumental line width. (b) Width corrected for instrumental effects and for assumed T dependence of shear viscosity mentioned in text. After Stinson and Litster.²⁷⁹

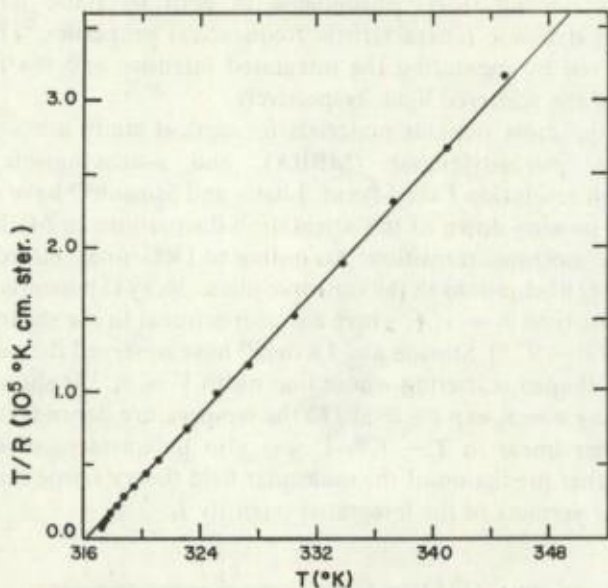


Fig. 15. Reciprocal intensity of the scattered light in isotropic MBBA vs. T . T_e is approximately 316°K. Solid line is a fit to mean field prediction.²⁸⁰

In the ordered phase a nematic liquid is conveniently described by a unit vector in the direction of molecular alignment $\hat{n}_0(r, t)$, often called the "director." Dynamical fluctuations of \hat{n}_0 are quite analogous to spin waves in a ferromagnet and have been studied in both PAA²⁷⁷ and MBBA.²⁸⁴ The PAA studies verified at constant temperature (125°C) that fluctuations δn_x can be decomposed into two uncoupled overdamped normal modes whose line widths are proportional to $a_i k^2$, where a_i depends on various geometry dependent combinations of the elastic and viscous coefficients of the nematic. Line widths (between 0.1 and 200 KHz) varied as k^2 for scattering angles between 1° and 40°. The MBBA experiments concentrated on the temperature dependence of both intensity and line widths for these modes observed at fixed k . On approaching T_c from below, definite line narrowing within $\sim 5^\circ$ of T_c and a slight increase in scattered intensity was observed.²⁸⁴ These results have been compared to a number of phenomenological theories for the dynamics of liquid crystals, but completely satisfactory agreement does not yet exist.

Some additional discrepancies with existing theory were found in the frequency dependent depolarization ratio for isotropic MBBA by Stinson, Litster, and Clark.²⁸⁰ This is shown for two temperatures in Fig. 16 together with theoretical predictions (solid curves). The phenomenological

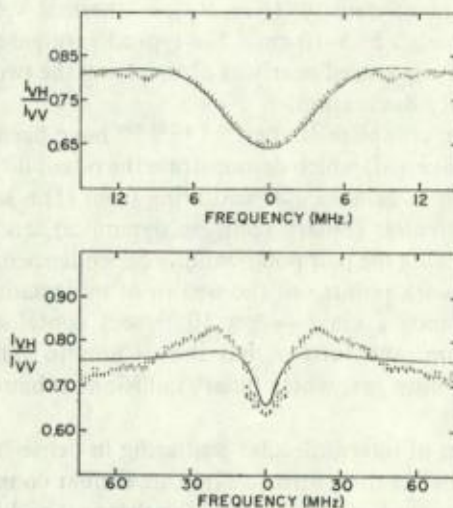


Fig. 16. Ratios of depolarized to polarized spectra in isotropic MBBA for $\theta = 6.55^\circ$. Upper curve and lower curves taken 10.54 and 24.14°C, respectively, above isotropic-nematic transition. Solid curves are best fit to phenomenological theory discussed by Stinson et al.²⁸⁰

theory of DeGennes ascribes the central dip to coupling between orientational fluctuations and hydrodynamic shear waves.²⁸⁰ In many ways the observations and explanations are similar to those discussed earlier for "shear waves" in normal anisotropic liquids, but a unified theoretical description covering both cases has yet to appear.

2. Intermolecular Light Scattering

A second important contribution to the spectra of all fluids was recently recognized for the first time. Since it arises from polarizability variations caused by *interactions among* fluid particles, we shall refer to it as "intermolecular light scattering." For dilute gases, however, the names "collision induced" and "translational Raman" scattering have often been applied. Traditionally, simple monatomic fluids were expected to exhibit no Raman or depolarized Rayleigh wing effects since they possess no vibrational or rotational degrees of freedom to modulate the polarizability. Levine and Birnbaum¹⁷⁶ first pointed out that a time-dependent polarizability results from the transient interaction between pairs of even the simplest atoms (i.e., binary collisions). The first observations of the spectra thus produced in dilute rare gases argon and krypton were made soon thereafter,^{178,179} which verified that the spectrum was (1) largely depolarized; (2) quasielastic of the form $I(\nu) = I_0 e^{-\nu/\Delta}$; with (3) $\Delta \simeq V_{TH}/r_0$ and Δ independent of density. (For V_{TH} = thermal velocity and r_0 = interaction distance, $\Delta \simeq 5\text{--}10\text{ cm}^{-1}$ for typical rare gases.) Also (4) the integrated intensity increased nearly as ρ^2 , implying the two-particle source of the polarizability fluctuation.

Since then more complete studies^{168,178,187,200} have been made of dilute rare gases (≤ 100 amagat) which demonstrate the possibility of quantitative calculation of their intermolecular scattering from (1) a knowledge of the intermolecular potential (binary collision dynamics), and (2) reasonable assumptions regarding the pair polarizability dependence on intermolecular separation. This work points out the wealth of information on the short-time dynamics (since $1\text{ cm}^{-1} \rightarrow 5 \times 10^{-12}$ sec) contained in the intermolecular spectrum, and further, has shown how to extract it explicitly for the case of a dilute gas, where binary collisions exhaust the short-time dynamics.

The observation of intermolecular scattering in dense fluids and liquids has stimulated considerable effort to explicate similar connections between the spectra and the short-time dynamics for these more difficult cases. The original experiments on liquid argon showed several interesting contrasts with the vapor at the same pressure and temperature.^{167,180} These experiments showed (i) the liquid spectrum is also nearly exponential in shape, (ii) Δ is nearly four times greater in the liquid than in the gas at the same

T , P , and (iii) the integrated intensity in the liquid is much smaller than expected from a density squared dependence. Thus, not surprisingly, the binary collision dynamics are demonstrably inadequate for liquids. McTague et al.¹⁸⁰ presented an interpretation of the liquid argon spectra based on a generalization of the familiar solid state concept of second-order Raman scattering.

Essentially a polarizability fluctuation of wave vector \mathbf{q} can be produced by a suitably paired set of fluctuations in some dynamic variable δU ; that is, $\delta\alpha_{\mathbf{q}} \sim \delta U_{-\mathbf{k}} \delta U_{\mathbf{k}+\mathbf{q}}$. δU may describe a fluctuation in vibration, rotation, magnetization, density, etc. Second-order Raman scattering in solids usually arises from pairs of vibrational fluctuations (phonons), predominantly those with $k \simeq \pi/a$, since the phonon density of states is largest near Brillouin zone boundaries. Halley¹⁹⁹ and Stephen²⁰¹ have applied similar ideas to the well-defined phonon-rotor dispersion curve in superfluid helium. Greytak and Yan¹⁹⁷ observed this second-order Raman scattering from superfluid helium, thus verifying the basic applicability of Stephen's theory (see Figs. 17 and 18). Stephen calculates the polarizability $\delta\alpha_{\mathbf{q}}$ produced by a density fluctuation $\delta\rho_{-\mathbf{k}}$ in the presence of the optical field and a second fluctuation $\delta\rho_{\mathbf{k}+\mathbf{q}}$. Recall from Section II that generally the scattered spectrum measures $\langle \delta\alpha_{\mathbf{q}}(t) \delta\alpha_{\mathbf{q}}^*(t + \tau) \rangle$, so that for this process it measures a fourth-order density correlation function. To the extent that interactions between the fluctuations are negligible this may be factored into $S(-\mathbf{k}, \Omega - \omega)S(\mathbf{k} + \mathbf{q}, \omega)$, where $S(q, \omega) = \langle \delta\rho_{-\mathbf{q}}(t = 0) \delta\rho_{\mathbf{q}}(\omega) \rangle$ is the familiar dynamic structure factor. For most solids and for superfluid helium, this situation obtains at least approximately. The intermolecular

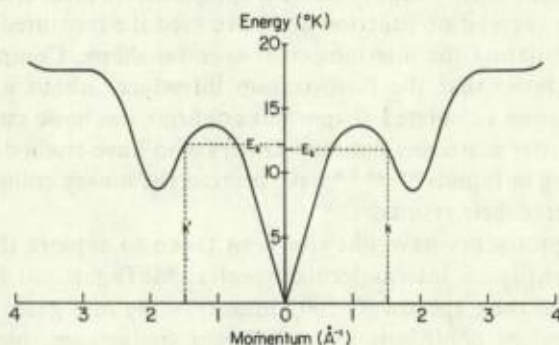


Fig. 17. Phonon-rotor dispersion curves in superfluid helium. Intermolecular scattering receives its dominant contributions from roton pairs whose individual momenta are near 1 \AA^{-1} and 2 \AA^{-1} where extrema occur in the dispersion curve. These should give peaks near 26°K and 18°K , respectively, in the roton pair spectrum. After Greytak and Yan.¹⁹⁷

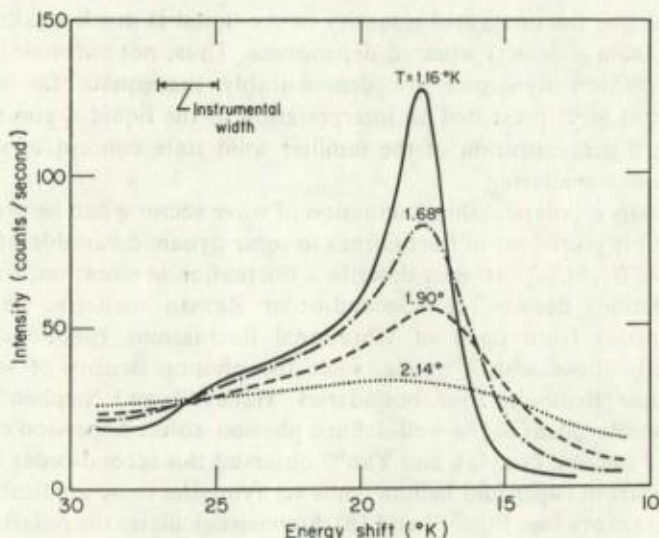


Fig. 18. Observed spectra in superfluid helium at various temperatures. The peak due to the roton minimum a $2 \times 9^\circ\text{K}$ is clear. The higher energy peak is suppressed by interaction effects. After Greytak and Yan.¹⁹⁷

spectrum can then be reasonably described by the factored correlation functions. (Note, however, that even small interaction effects have important implications for the roton spectra.¹⁹⁸) For a classical liquid like argon, McTague et al.¹⁸⁰ argued that the spectrum still measures the fourth-order density correlation function and have used the measured $S(\mathbf{k}, \omega)$ for argon in calculating the intermolecular spectral shape. Comparison with experiment shows that the factorization introduces about a 25% error into the resulting calculated shape, but confirms the basic correctness of the second-order scattering concept. Others who have studied intermolecular scattering in liquids^{169,163,186} have utilized the binary collision formalism to interpret their results.

Various approaches have likewise been taken to explore the effects of increasing density on intermolecular spectra. McTague and Birnbaum¹⁷⁹ have described their spectra (0–200 amagat) in the rare gases in terms of density-dependent contributions at different frequencies, identifying ρ^n components with n -particle collisions, etc. Fleury et al.¹⁶⁶ have measured the intermolecular spectra of argon and neon as functions of temperature and density over a continuous range extending from the dilute gas limit to densities as high as 50% in excess of the normal liquid density. They were thus able to follow the evolution of the spectra throughout these ranges. Typical results for argon are shown in Fig. 19, where it can be seen that

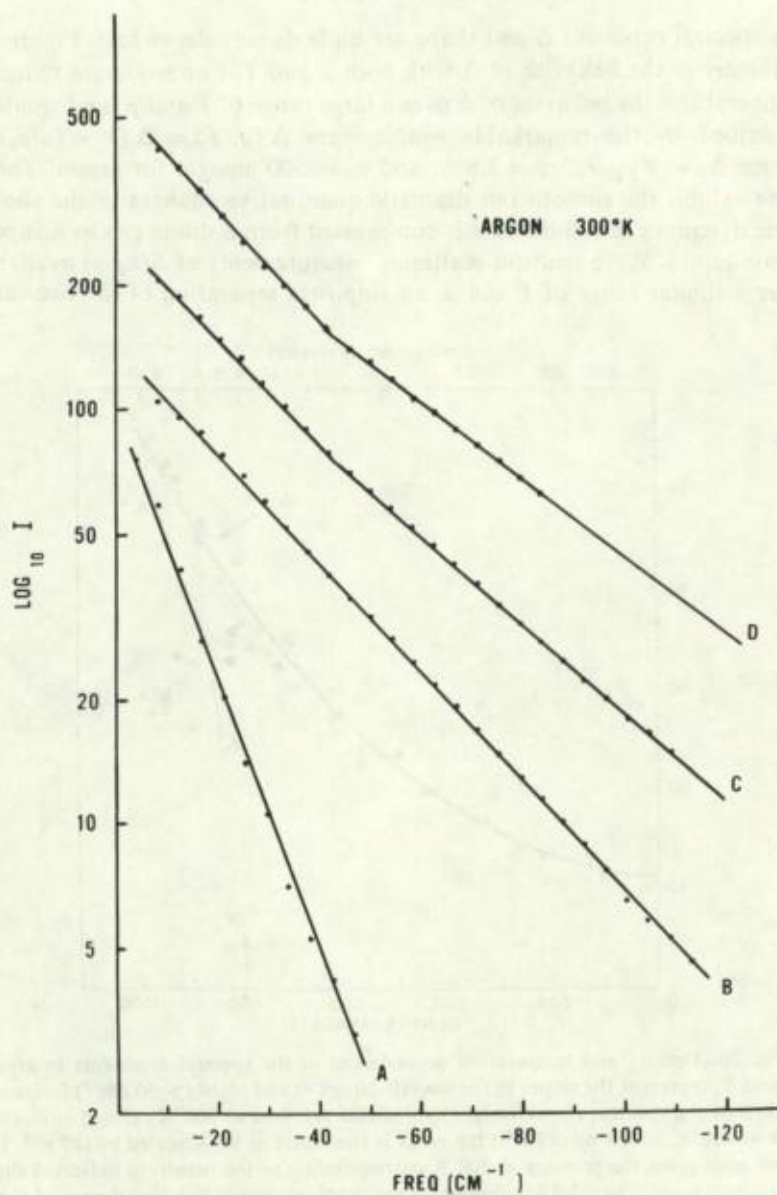


Fig. 19. Depolarized Stokes spectra for argon at 300°K for different densities. For display the curves are arbitrarily displaced along the vertical scale. The slopes of the straight lines drawn through the data points give the values of Δ plotted in Fig. 20. Curve A, 200; curve B, 750; curve C, 825; and curve D, 905 amagats. Note log scale for intensity. After Fleury et al.¹⁰⁶

the spectral exponent Δ and shape are quite density dependent. Figure 20 summarizes the behavior of Δ with both ρ and T . For moderate reduced temperatures the behavior of Δ over a large range of T and ρ is adequately described by the remarkably simple form $\Delta(\rho, T) = \Delta_0(1 + (\rho/\rho_0)^2)$, where $\Delta_0 = V_{TH}/r_0$, $r_0 = 2.8 \text{ \AA}$, and $\rho_0 = 500 \text{ amagat}$ for argon. These data exhibit the smooth but dramatic quantitative changes in the short-time dynamics of a fluid as it is compressed from a dilute gas to a superdense liquid. Were neutron scattering measurements of $S(k, \omega)$ available over a similar range of T and ρ , an empirical separation of the two- and

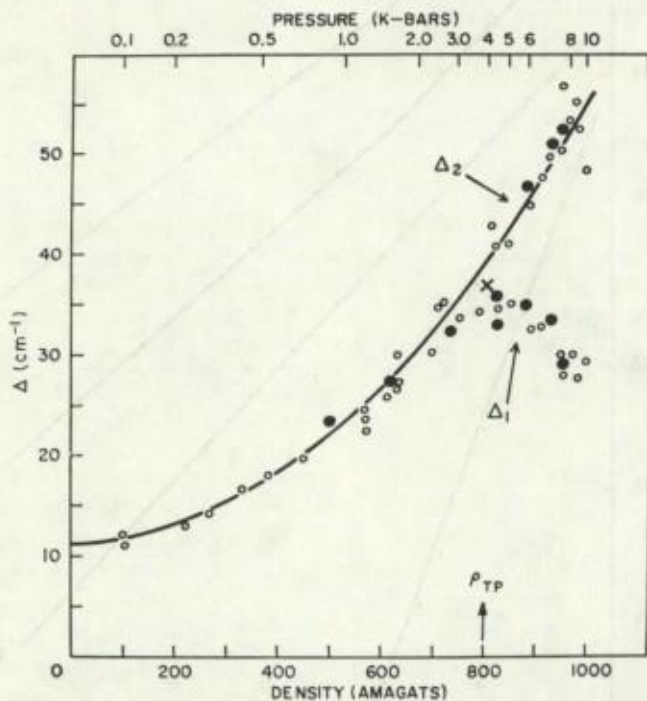


Fig. 20. Density and temperature dependences of the spectral exponents in argon. Δ_1 and Δ_2 represent the slopes in the low- ($0-50 \text{ cm}^{-1}$) and high- ($> 50 \text{ cm}^{-1}$) frequency regions of the spectra, respectively. Open circles are data at 300°K ; closed circles are data at 180°K scaled by $(kT)^{1/2}$; the cross is the liquid at 90°K scaled by $(kT)^{1/2}$. The upper scale gives the pressure at 300°K corresponding to the density ρ indicated along the bottom scale. The solid line represents the simple empirical function discussed in the text. After Fleury et al.¹⁶⁶

four-point density correlation functions would be possible. In their absence we must rely on developing theories of the dynamics of dense fluids to provide the quantitative connection between intermolecular spectra and the microscopic dynamic fluid structure. Despite the embryonic nature of our present understanding, it seems likely that further cooperative theoretical and experimental work on intermolecular light scattering will contribute significantly to a successful theory of the liquid state.

Finally, the intermolecular spectra of both He³ and He⁴ have been observed by Slusher and Surko¹⁸⁴ under pressure up to 150 atm. In the normal fluid ranges they are qualitatively similar to those observed in the classical fluids. The same workers have verified the second-order Raman effect in the solid phases of He³ and He⁴. As with the other rare gas solids, the spectral shapes change surprisingly little upon solidification.

E. Surface Scattering

Over half a century elapsed between the realization that light should be scattered by a surface and the measurement of the spectral content of that scattered light.^{207,208} Thermally excited capillary waves or "ripples" are describable through a rather complicated dispersion relation^{203,214} whose essential physical content we now discuss. Surface tension σ provides the restoring force, and shear viscosity η_s the major contribution to damping for these waves. For wave vectors $k > k_c \equiv 2\pi\sigma\rho/3.65\eta_s^2$, the ripples are overdamped. There is a maximum frequency for surface wave propagation $\omega_m = 0.375\sigma^2\rho/\eta_s^3$ regardless of wave vector. These waves, of course, are also sustained at the interface between two fluids and exhibit quite interesting behavior near the liquid-vapor critical point or near the critical mixing point. In particular, several experiments on liquid-gas interface which measure the critical slowing down of the surface waves near C.P. have now been reported.²¹³⁻²¹⁵ This slowing down is essentially a result of the vanishing of the surface tension σ as $T \rightarrow T_c$. Typical k values explored in these experiments are $\ll k_c$, so that the ripples are not overdamped. Although generally rather complicated, the dispersion relation for waves on the interface between two fluids of densities ρ_1 and ρ_2 and viscosities η_1 and η_2 can be approximately written (in the incompressible limit²¹⁴)

$$y = -\frac{2\rho_1\rho_2}{(\rho_1 + \rho_2)^2} S(1 + 2S)^{1/2} [1 + (1 + 2S)^{1/2}] \\ - \frac{(\rho_1 - \rho_2)^2}{(\rho_1 + \rho_2)^2} [(1 + S)^2 - (1 + 2S)^{1/2}] \quad (100)$$

where

$$y = \frac{[\sigma k^3 + g(\rho_1 - \rho_2)][\rho_1 + \rho_2]}{4k^3(\eta_1 + \eta_2)^2} \quad (101)$$

and

$$S = (-\Gamma + i\omega) \frac{\rho_1 + \rho_2}{2k^2(\eta_1 + \eta_2)} \quad (102)$$

ω and Γ thus give the frequency and damping of the ripples. Typically $\omega \sim 5$ KHz and $\Gamma \sim 1$ KHz for $k \simeq 2000 \text{ cm}^{-1}$, σ of ~ 0.1 dyne/cm, and normal liquid ρ 's and η 's. Thus light beating spectroscopy has usually been employed to study them.

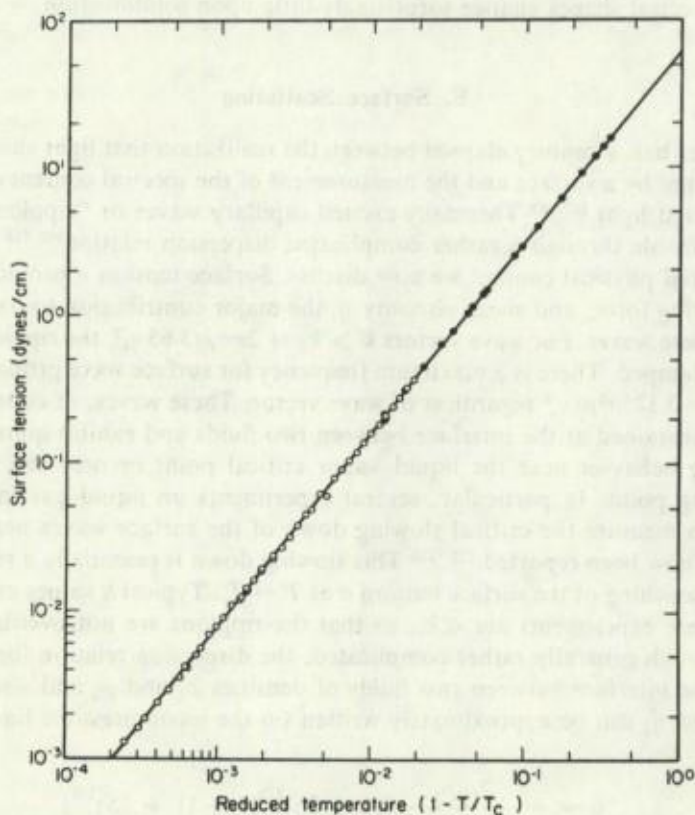


Fig. 21. Surface tension of xenon as a function of reduced temperature $1 - T/T_c$. Open circles inferred from surface wave scattering spectra. Solid circles from conventional capillary rise experiments. After Zollweg et al.²¹³

By analyzing the k and T dependence of the ripplon spectra, one may deduce both $(\rho_1 - \rho_2)$, the C.P. order parameter, and σ , the vanishing surface tension. In fact, light scattering has extended by orders of magnitude the range of σ 's that can be measured. Figure 21 shows for xenon both light scattering and conventional capillary rise data for σ over several decades in ϵ .²¹³ σ obeys a power law in ϵ over a remarkable range with $(\sigma = \sigma_0 \epsilon^\mu)$ a value of $\mu = 1.302 \pm 0.006$. Similar experiments on CO_2 and SF_6 have produced μ 's of 1.25 ± 0.01 and 1.30 ± 0.07 , respectively.^{214,215} Ripplon spectra also contain information on the correlation length, ξ , and on the closely related interfacial thickness L , both of which diverge at C.P.²¹³

Langevin and Bouchiat²⁶² have studied surface wave scattering in the nematic liquid crystals MBBA and PAA and have deduced the temperature dependences of the *three* independent shear viscosity parameters as well as the surface tension σ near the isotropic-nematic transition.

Clearly the field of two-dimensional fluid behavior as viewed through surface and thin film^{212,205,273} waves is just opening up and we can expect light scattering to play an important role in its promising future.

F. Solutions and Macromolecules

The simplest dynamic information obtainable in a light scattering experiment is the uncorrelated motion of single polarizable particles. In very dilute gases the particle motions obey a Maxwell-Boltzmann velocity distribution which produces an appropriately Doppler broadened (Gaussian) frequency profile in the scattered light. If these polarizable particles are embedded in a nonpolarizable fluid with which they make frequent collisions, they will still scatter light but with a different frequency profile (Lorentzian) which describes their diffusive or Brownian motion. The line width $\Gamma = Dk^2$ then measures the translational mass diffusion coefficient of the particles in the surrounding medium.³⁰⁷ Macromolecules of biological interest exhibit D 's of order 10^{-6} cm²/sec, producing Γ 's in the KHz range for visible light scattering ($k \approx 10^5$ cm⁻¹). This fact and the rapid development of electronic spectrometers has made the light scattering technique a standard one for rapid determination of diffusion coefficients, particularly in dilute solutions ($\sim 0.01\%$). However, since most interesting molecules are anisotropic, and often long enough to contain segments comparable to optical wavelengths, the above simple picture of translational diffusion is almost always inadequate. In addition even the translational diffusion coefficient may be different for motion parallel (D_{\parallel}) and perpendicular (D_{\perp}) to the long molecular axis. Further, the molecules may

interact with each other or with a charge compensating cloud of counterions arising from any charge which may reside on the dissolved macromolecules. All these effects greatly complicate the light scattering spectra in real systems. If the solute molecules also react or polymerize, or change conformation, further spectral complexity results from the accompanying polarizability modulations and/or changes in constituent mass, volume, or shape. Although, in principle, each of these effects produces a calculable change in the observed spectrum, not enough is known at present about the values of the necessary parameters to infer uniquely their separate contributions from a given set of experimental spectra. Nevertheless the considerable experimental and theoretical effort now being expended on such complications is cause for optimism, and even in the absence of perfectly quantitative spectral interpretation, considerable valuable information has already been obtained from light scattering experiments on solutions of macromolecules.

In view of the recent review article on this subject by Pecora,¹⁴ our discussion here will be very brief. Most experiments^{316,361} have deduced the translation D_T and rotational D_R diffusion coefficients from the spectral line shapes. If these diffusion processes are uncoupled, they can be inferred from the polarized (I_{zz}) and depolarized (I_{zy}) line shapes¹⁴:

$$I_{zz}(\omega, k) = \frac{A}{9\pi} (\alpha_{\parallel} + \alpha_{\perp})^2 \frac{k^2 D_T}{\omega^2 + (k^2 D_T)^2} + \frac{4}{3} I_{zy}(\omega, k)$$

$$I_{zy}(\omega, k) = \frac{A}{15\pi} (\alpha_{\parallel} - \alpha_{\perp})^2 \frac{k^2 D_T + L^{-2} D_R}{\omega^2 + (k^2 D_T + L^{-2} D_R)^2}$$

where L = molecule rod length; α_{\parallel} , α_{\perp} molecular polarizabilities; A = Const. Several workers^{316,361} have used these formulas to describe their data on solutions of tobacco mosaic virus (TMV). Schaeffer et al. have derived more complicated expressions which additionally account for anisotropic translational diffusion to apply to their TMV results.³⁶³ They also mention possible complications arising from counterion clouds interacting with the macromolecules, but conclude these complications do not apply to their experiments. It would be of considerable interest, however, to observe these effects experimentally. Stephen has shown that counterion association will result in an enhanced apparent $D_T' = D_T(1 + \alpha)$ under certain circumstances.³⁴³ (α depends on the Debye screening length of the macromolecular charge relative to the scattering wave vector, k .) This effect provides in principle a means to measure residual charge on macromolecules in solution.

Diffusion measurements on DNA,³⁶² R-17 virus, lysozyme, myosin, and *E. coli* bacteria have also been reported.^{356,359}

Recently some intriguing studies of the nonequilibrium motion of bacteria by light scattering have been done by Nossal and Chen.³⁵⁹ Such techniques provide exciting possibilities for microscopic studies of chemotaxis, bacterial association, and related phenomena. Indeed these experiments are very similar in technique and spirit to the Doppler shifted fluid flow measurements carried out by hydrodynamicists and plasma physicists.³¹³

A final remark about molecular solutions should be made. It has been pointed out by several authors that light will be inelastically scattered by the concentration fluctuations associated with formation and destruction of components participating in a chemical reaction.^{286-289,324} In the simplest case of optically isotropic constituents reacting at a rate $K = k_{12} + k_{21}$ much greater than the diffusion rate, $k^2 \bar{D}_T$, the polarized spectrum has the simple form¹⁴:

$$I_{zz} = C |\bar{\alpha}|^2 \frac{k^2 \bar{D}_T}{\omega^2 + (k^2 \bar{D}_T)^2} + \frac{C_1 C_2}{C} |\alpha_1 - \alpha_2|^2 \frac{K}{\omega^2 + K^2}$$

where α_i and C_i are polarizability and equilibrium concentrations of species i ; and $\bar{\alpha} = (1/C)[C_1 \alpha_1 + C_2 \alpha_2]$; $C = C_1 + C_2$.

The dearth of experimental observations of chemical reaction scattering stems from the scarcity of simple systems^{346,347} having α_1 much different from α_2 and C_1, C_2 both of order C . This scarcity somewhat attenuates the initial attractiveness of light scattering for studying fast chemical reactions. (Rates $K \geq 10^{10} \text{ sec}^{-1}$, for example, would be easily studied with Fabry Perot techniques.)

As with the film and surface wave studies mentioned above, the field of light scattering from chemical reactions and macromolecular solutions has a promising future.

Literature Search and Bibliography

This section consists of a relatively complete list of references, including titles on the general subject of inelastic light scattering in fluids. The list is a result of a literature search, conducted with the assistance of Bernard Stevens of Bell Laboratories, concentrating on the period beginning with the introduction of lasers to spectroscopy (1964) to approximately the beginning of 1972. Because no truly centralized and complete collection of all physics, chemistry, and biology literature exists, even for this recent period, the bibliography cannot be absolutely complete. However, it is the most extensive one yet compiled on the subject and will hopefully prove useful to both students and researchers.

The bibliography is arranged as follows: papers are divided into twelve categories. In addition to the reference information, the title of each paper

is cited in full. In the event that a paper may fall into more than one category, it is listed under the one we judge the most applicable. To avoid having to duplicate those papers actually cited in the text, all papers whose primary content places them in a given category are numbered consecutively beginning with the first in category 1 until the last in category 12. These consecutive numbers are also used to designate textual citations when appropriate. Although this results in the textual references not being numbered in the order of their citation, we feel it is the most concise way of having the bibliography serve two purposes without introducing unnecessary confusion. Within the categories papers are listed alphabetically by first author. A few papers appear in more than one category but are numbered only when appearing in their primary category. Their other listings are unnumbered, but the primary category to which they belong is then indicated by the number in brackets at the end of each such entry.

As mentioned in the introduction, the subjects of ordinary Raman scattering, scattering in solids and plasmas, and stimulated scattering were defined to be outside the scope of this review. With the exceptions of the latter topic, these exclusions apply as well to the bibliography. Nevertheless, we include stimulated scattering in the bibliography primarily because such a large number of references were revealed in the search that it was deemed useful to present them in a concise collection.

The categories defined are as follows, (1) review articles or books on various aspects of light scattering in fluids; (2) instrumentation and experimental technique; (3) Rayleigh scattering from entropy fluctuations; (4) Brillouin scattering; (5) intermolecular or collision induced scattering; (6) scattering from surfaces and thin films; (7) molecular anisotropy and orientation effects; (8) liquid crystals; (9) mixtures and particles in solution; (10) critical phenomena; (11) stimulated scattering; and (12) nonlinear scattering and miscellaneous references.

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