A CORRELATION THEORY OF RAYLEIGH SCATTERING

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We use relaxation theory [5] to establish the form of the dispersion laws for the complex coefficients referring to the compression waves, which occur in the general spectral equations of the dynamics and heat transfer in an isotropic medium.[1] We find a connection between the dispersion laws of three coefficients: the compression modulus $\overline{K}$, the thermal expansion coefficient $\overline{\alpha}$, and the specific heat $\overline{c}_v$ for the case when the compression waves have a single relaxation time (Eq. (11)). As an illustration of the limitations imposed by Eq. (11) when phenomenological dispersion laws are given we consider three models of a medium, used in the literature.[3,4,6] We discuss the influence of the choice of model upon the spectrum of light scattered by density fluctuations. In the theory of scattering developed by Mountain [6,7] an error is shown to occur and as a result it describes scattering in the liquid with a dispersion only in the shear modulus, notwithstanding the original statement of the problem.

1. STATEMENT OF THE PROBLEM

THE set of linearized spectral equations for the displacement s and velocity v in an visco-elastic medium and for the deviations from the equilibrium values of the specific entropy ($S_1 = S - S_0$), density ($\rho_1 = \rho - \rho_0$), and temperature ($T_1 = T - T_0$) can be written in the form

$$i\omega\rho_0 v_\alpha = \frac{\partial\sigma_{\alpha\beta}}{\partial x_\beta}, \quad i\omega S_1 = \frac{\varkappa}{\rho_0 T_0}\nabla^2 T_1, \quad S_1 = \frac{\overline{\alpha}}{3\rho_0}\sigma + \bar{c}_p\frac{T_1}{T_0}. \tag{1}$$

$$v_\alpha = i\omega s_\alpha, \quad \sigma_{\alpha\beta} = 2\bar{\mu}\tilde{u}_{\alpha\beta} + \overline{K}(u - \overline{\alpha}T_1)\delta_{\alpha\beta},$$

$$u_{\alpha\beta} = \frac{1}{2}\left(\frac{\partial s_\alpha}{\partial x_\beta} + \frac{\partial s_\beta}{\partial x_\alpha}\right), \quad \tilde{u}_{\alpha\beta} = u_{\alpha\beta} - \frac{u}{3}\delta_{\alpha\beta}, \tag{2}$$

$$u \equiv u_{\alpha\alpha} = \partial s_\alpha/\partial x_\alpha = -\rho_1/\rho_0, \quad \sigma \equiv \sigma_{\alpha\alpha} = 3\overline{K}(u - \overline{\alpha}T_1).$$

Here $\sigma_{\alpha\beta}$ and $u_{\alpha\beta}$ are, respectively, the tension and deformation tensors, κ the thermal conductivity coefficient, and the four quantities with a bar on top are complex functions of the frequency ω which we can conveniently call by their usual name although, strictly speaking, the usual nomenclature refers only to a quasi-equilibrium state ($\omega \to 0$). In fact, $\overline{K}$ and $\bar{\mu}$ are complex elastic moduli:

$$\overline{K}(i\omega) = K(\omega^2) + i\omega\zeta(\omega^2), \quad \bar{\mu}(i\omega) = \mu(\omega^2) + i\omega\eta(\omega^2),$$

i.e., K is the "isothermal" hydrostatic compression, and μ the shear modulus; ζ and η are the bulk and shear viscosities; $\overline{\alpha}$ and $\overline{c}_p$ are the complex thermal expansion coefficient and specific heat "at constant pressure." In the following it will be convenient to introduce three more complex quantities: the specific heat "at constant volume" $\overline{c}_v$, the Poisson ratio γ, and the "adiabatic" compression ratio $\overline{K}_a$, which are equal to

$$\bar{c}_v = \bar{c}_p - \frac{T_0}{\rho_0}\overline{\alpha}^2\overline{K}, \quad \overline{\gamma} = \frac{\bar{c}_p}{\bar{c}_v}, \quad \overline{K}_a = \overline{\gamma}\overline{K}. \tag{3}$$

When $\omega = 0$, all these parameters take on the real thermodynamic values which we shall indicate by an index 0 ($K(0) = K_0$, $\mu(0) = \mu_0$, and so on; for liquids and gases $\mu_0 = 0$).

When there is no spatial dispersion, Eqs. (1) and (2) which satisfy the symmetry conditions of kinetic coefficients and which as $\omega \to 0$ change to the usual equations for a non-dissipative medium, give the most general spectral description of a visco-elastic isotropic continuous medium. In a somewhat different notation these equations have been used to study equilibrium thermal fluctuations in such a medium with an arbitrary frequency dispersion of the parameters [1] and this made it possible to obtain afterwards general spectral formulae for the Rayleigh scattering of light in such a medium.[2,3]

Of course, the results of such a phenomenological theory must be made concrete whenever one deals with a comparison with experiments, for instance, for dispersion and damping of ultrasound, the spectral distribution of the intensity of scattered light, or the integral values of the intensities of the different components of this light. This means that the dispersion laws of the coefficients $\overline{K}$, $\overline{\alpha}$, $\overline{c}_p$, and $\bar{\mu}$ must be introduced externally, i.e., they must be taken either from a well-defined model of the medium as, for instance, in Kneser's theory for diatomic gases [4] or from a general "thermodynamic" theory which takes into account the kinetics of some internal parameters as was done by Mandel'shtam and Leontovich for compression waves in liquids.[5]

However, in many papers the dispersion laws for the coefficients of the spectral equations are given formally satisfying only the condition that they do not violate the dissipativeness of the medium and the problem then arises in how far these laws are independent of one another. The present paper is also devoted to some considerations in this connection and some of the consequences arising from them mainly for the theory of Rayleigh scattering. In the concluding section we give an analysis of the scattering theory in liquids which was recently developed by Mountain.[6,7]

2. DISPERSION OF COMPRESSION WAVES

In accordance with the basic ideas of [5], Eqs. (1) and (2) are a consequence of some more general set of linear equations which are satisfied not only by the mechanical and thermal variables s, v, $u_{\alpha\beta}$, $\sigma_{\alpha\beta}$, S_1, and T_1, but also by some internal parameters ξ_k ($k = 1,\ldots,n$). One obtains Eqs. (1) and (2) by eliminating the ξ_k and hence the dispersion of the coefficients in (1) and (2) is determined by the kinetics of the same parameters ξ_k. It is clear that when the number of these hidden parameters n is sufficiently large one can always choose such a model of the medium that with the accuracy required practically any independently given (but physically realized) frequency dependencies of the quantities $\overline{K}$, $\overline{\alpha}$, $\overline{c}_p$, and $\overline{\mu}$ can be reproduced. It is sufficient to appeal to the case, studied in[5], of a liquid with such a small shear viscosity and thermal conductivity that we can completely neglect them ($\overline{\mu} = 0$, $\kappa = 0$) thus assuming that the propagation of the propagation of the compression waves is isentropic.

The "reaction equations," i.e., the kinetic equations for $\xi = \{\xi_1,\ldots,\xi_n\}$ were in[5] written in the following spectral form:

$$(1 + i\omega\tau_k)\xi_k = E_k\frac{T_1}{T_0} - P_k\frac{\rho_1}{\rho_0^2}, \tag{4}$$

where $E = E(T, \rho, \xi)$ is the internal energy per unit mass of the liquid, $p = P(T, \rho, \xi)$ the pressure, $E_k = (\partial E/\partial\xi_k)_0$, $P_k = (\partial P/\partial\xi_k)_0$, and τ_k the relaxation time of the parameter ξ_k. The energy equation following from the condition of isentropic behavior has the form

$$S_1 = C_v\frac{T_1}{T_0} - P_T\frac{\rho_1}{\rho_0^2} + \frac{1}{T_0}\sum_{k=1}^{n} E_k\xi_k = 0. \tag{5}$$

Capital letters indicate derivatives for ξ_k = const:

$$C_v = \left(\frac{\partial E}{\partial T}\right), \qquad P_T = \left(\frac{\partial P}{\partial T}\right)_\xi,$$

in contrast to the derivatives corresponding to the equilibrium state, i.e., taken for $\nu_k \equiv \partial\Psi(T, \rho, \xi)/\partial\xi_k$ = const (Ψ: free energy):

$$c_v = \left(\frac{\partial E}{\partial T}\right)_\nu, \qquad p_T = \left(\frac{\partial P}{\partial T}\right)_\nu.$$

Eliminating the ξ_k from (5) by means of (4) we get an expression for S_1 in terms of T_1 and ρ_1. Comparison with the formula for S_1 following from (1) and (2) gives the dispersion laws for the specific heat $\overline{c}_v$ and the product $\overline{\alpha}\overline{K}$.[1)] Moreover, the expression obtained in[5] for the complex velocity of propagation of the compression waves $V(i\omega)$ immediately determines the "adiabatic" $\overline{K}_\alpha = \gamma\overline{K}$ since for $\overline{\mu} = 0$ and $\kappa = 0$ we have $V^2 = \overline{K}_a/\rho_0$. As a result of those comparisons we have three formulae:

$$\bar{c}_v = C_v + \frac{R_1(i\omega)}{T_0}, \qquad \overline{\alpha}\overline{K} = P_T + \frac{R_2(i\omega)}{T_0},$$

$$\overline{K}_\alpha = \overline{\gamma}\overline{K} = \rho_0 P_\rho - \frac{R^3(i\omega)}{\rho_0} + \frac{T_0(\overline{\alpha}\overline{K})^2}{\rho_0\bar{c}_v}, \tag{6}$$

where we have introduced the notation[2)]

$$R_1(i\omega) = \sum_{k=1}^{n}\frac{E_k^2}{1 + i\omega\tau_k}, \qquad R_2(i\omega) = \sum_{k=1}^{n}\frac{P_kE_k}{1 + i\omega\tau_k},$$
$$R_3(i\omega) = \sum_{k=1}^{n}\frac{P_k^2}{1 + i\omega\tau_k}. \tag{7}$$

to simplify the equations. It follows from (6) and (3) that the dispersion laws for the quantities $\overline{K}$, $\overline{\alpha}$, and $\overline{c}_p$ have the form

$$\overline{K} = \rho_0 P_\rho - \frac{R_3(i\omega)}{\rho_0}, \quad \overline{\alpha} = \frac{1}{\overline{K}}\left(P_T + \frac{R_2(i\omega)}{T_0}\right),$$
$$\bar{c}_p = C_v + \frac{R_1(i\omega)}{T_0} + \frac{T_0}{\rho_0}\overline{\alpha}^2\overline{K}, \tag{8}$$

and the total dispersion drops (from $\omega = 0$ to $\omega = \infty$) of the quantities $\overline{K}$, $\overline{\alpha}\overline{K}$, and $\overline{c}_v$ are according to (6) to (8) given by

$$\Delta K = K_\infty - K_0 = \frac{R_3(0)}{\rho_0} = \frac{1}{\rho_0}\sum_{k=1}^{n} P_k^2,$$
$$\Delta(\alpha K) = \alpha_\infty K_\infty - \alpha_0 K_0 = -\frac{R_2(0)}{T_0} = -\frac{1}{T_0}\sum_{k=1}^{n} P_kE_k, \tag{9}$$
$$\Delta c_v = c_{v\infty} - c_{v0} = -\frac{R_1(0)}{T_0} = -\frac{1}{T_0}\sum_{k=1}^{n} E_k^2.$$

Comparing these expressions with the ones derived in[5] from the relations between derivatives with ξ = const and with ν = const we see that $K_0 = \rho_\sigma p_\rho$ $\alpha_0 K_0 = p_T$ and $c_{v\infty} = C_v$.

The frequency dependence of the three coefficients $\overline{K}$, $\overline{\alpha}$, and $\overline{c}_p$ which remain in Eqs. (1) and (2) when $\overline{\mu} = 0$ is thus in final reckoning determined by the values of the 3n quantities E_k, P_k, and τ_k ($k = 1,\ldots,n$) and the total dispersion drops of these coefficients by the three quantities: $R_1(0)$, $R_2(0)$, and $R_3(0)$ which depend only on the E_k and the P_k. The statement made earlier about the possible reproduction of any independently given dispersion laws for $\overline{K}$, $\overline{\alpha}$, and $\overline{c}_p$ for sufficiently large n is obvious. Already in the particular case of Kneser's theory when the ξ_k are the concentrations of excited molecules and the pressure is independent of the ξ_k ($P_k = 0$, $R_2 = R_3 = 0$) we have $K = K_0$, $\alpha = \alpha_0$ but for the approximation of any dispersion law for $\overline{c}_p$ (or $\overline{c}_v$) there remain in our arrangement 2n quantities E_k and τ_k.

The position changes appreciably if we introduce the often used assumption about the presence of only a single relaxation time for the quantities which are directly connected with compression waves; we shall denote it by τ'. Equations (7) in which now n = 1 given then

$$R_1(i\omega) = \frac{E_\xi^2}{1 + i\omega\tau'}, \quad R_2(i\omega) = \frac{E_\xi P_\xi}{1 + i\omega\tau'}, \quad R_3(i\omega) = \frac{P_\xi^2}{1 + i\omega\tau'} \tag{10}$$

and hence <u>for any</u> τ' we have the relation $R_2^2 = R_1R_3$ from which follows in accordance with (6) and (8) that

$$T_0(\alpha_\infty K_\infty - \alpha\overline{K})^2 = -\rho_0(K_\infty - \overline{K})(c_{v\infty} - \bar{c}_v). \tag{11}$$

In particular, putting here $\omega = 0$ we get a relation between the total dispersion drops:

$$T_0[\Delta(\alpha K)]^2 = -\rho_0\Delta K\Delta c_v. \tag{12}$$

In the case of a single (arbitrary) relaxation time the dispersion laws for the coefficients $\overline{K}$, $\overline{\alpha}$, and $\overline{c}_v$

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1)For the mentioned comparison it is clearly unimportant that in[5] it was assumed that $S_1 = 0$.

2)We note that for the real and imaginary parts of $R_k = R_k' - iR_k''$ we have the obvious inequalities

$$h^2R_1' + R_3' \pm 2hR_2' \geqslant 0, \qquad h^2R_1'' + R_3'' \pm 2hR_2'' \geqslant 0$$

(h is a factor with the dimensions of a density) from which one can derive some limitations for the real and imaginary parts of $\overline{K}$, $\overline{\alpha}\overline{K}$, and $\overline{c}_v$.

(or $\bar{c}_p$) are thus connected. Assuming, e.g., that $\Delta K = 0$ we must by virtue of (12) assume also $\Delta\alpha = 0$, and when $\Delta c_V = 0$, $\Delta(\alpha K) = 0$ is also necessary.

3. MEDIUM WITH WEAK DISPERSION

In contrast with the preceding section we shall now take the shear modulus $\bar{\mu}$ and the thermal conductivity κ into account, but we shall consider only acoustic and thermal waves. To get rid of the shear waves it is sufficient to put $\mathbf{v} = \operatorname{grad}\varphi$ and then elimination of all variables bar the velocity potential φ and the temperature fluctuations T_1 from the set (1) and (2) leads to the equations

$$\begin{gathered}\omega^2\rho_0\varphi + (\bar{K} + {}^4/_3\bar{\mu})\nabla^2\varphi - \bar{\alpha}\bar{K}i\omega T_1 = 0,\\ \bar{\alpha}\bar{K}T_0\nabla^2\varphi + \rho_0\bar{c}_v i\omega T_1 - \varkappa\nabla^2 T_1 = 0.\end{gathered} \tag{13}$$

We restrict ourselves to weakly dispersive media for which we can retain in all formulae only terms in first order in the disperion drops and also in the thermal conductivity κ. If we use Eqs. (9) we obtain from Eqs. (6) and (8) the following dispersion laws for $\bar{K}$, $\bar{\alpha}$, and $\bar{c}_V$ and for the specific heat $\bar{c}_p$ which is connected with them through the first of Eqs. (3):

$$\begin{gathered}\bar{K} = K_0\Big(1 + \frac{\Delta K}{K_0}\psi_3\Big), \qquad \bar{c}_v = c_{v0}\Big(1 + \frac{\Delta c_v}{c_{v0}}\psi_1\Big),\\ \bar{\alpha} = \alpha_0\Big[1 + \frac{\Delta\alpha}{\alpha_0}\psi_2 + \frac{\Delta K}{K_0}(\psi_2 - \psi_3)\Big],\\ \bar{c}_p = c_{p0}\Big\{1 + \frac{\Delta c_p}{c_{p0}}\psi_1 + \Big(1 - \frac{1}{\gamma_0}\Big)\Big[\frac{\Delta K}{K_0}(2\psi_2 - \psi_1 - \psi_3)\\ + 2\frac{\Delta\alpha}{\alpha_0}(\psi_2 - \psi_1)\Big]\Big\}.\end{gathered} \tag{14}$$

Here

$$\psi_k = 1 - \frac{R_k(i\omega)}{R_k(0)} \qquad (k = 1, 2, 3) \tag{15}$$

and we have introduced the relative drop $\Delta c_p/c_{p0}$ for which we have from (3) in first order the expression

$$\frac{\Delta c_p}{c_{p0}} = \frac{\Delta c_v}{\gamma_0 c_{v0}} + \Big(1 - \frac{1}{\gamma_0}\Big)\Big(\frac{\Delta K}{K_0} + 2\frac{\Delta\alpha}{\alpha_0}\Big) \tag{16}$$

With the same accuracy the condition (12) changes in the case of a weakly dispersive medium to the following one:

$$\frac{\Delta K}{K_0}\frac{\Delta c_v}{c_{v0}} = -(\gamma_0 - 1)\Big(\frac{\Delta K}{K_0} + \frac{\Delta\alpha}{\alpha_0}\Big)^2. \tag{17}$$

Although now $\bar{\mu} \neq 0$ we proceed nevertheless from the earlier formulae for the quantities which describe the bulk elasticity of the medium i.e., we assume that there is no interdependence of the dispersion laws for $\bar{K}$, $\bar{\alpha}$, and $\bar{c}_V$, on the one hand, and the shear modulus $\bar{\mu}$, on the other hand. This is correct because $\bar{\mu}$ characterizes the elasticity of the medium for deformations or motions of a completely different (vortex) kind while the relaxation times for compression and shear are usually different.

We now turn to the simplest case when for each type of wave we assume the existence of a single relaxation time τ' for the quantities $\bar{K}$, $\bar{\alpha}$, and $\bar{c}_V$ and τ for the shear modulus $\bar{\mu}$:

$$\bar{\mu} = \mu + i\omega\eta = \frac{\mu_0 + i\omega\tau\mu_\infty}{1 + i\omega\tau}. \tag{18}$$

For low-viscosity liquids usually $\tau \ll \tau'$ i.e., the dispersion of the compression is completed at much lower frequencies than the dispersion of the shear starts to appear. According to (10) and (15)

$$\psi_1 = \psi_2 = \psi_3 = \frac{i\omega\tau'}{1 + i\omega\tau'} \equiv \psi \tag{19}$$

and the dispersion formulae (14) take the same form

$$\bar{A} = A_0\Big(1 + \frac{\Delta A}{A_0}\psi\Big), \tag{20}$$

where for $\bar{A}$ we can take any of the quantities $\bar{K}$, $\bar{\alpha}$, $\bar{c}_V$, $\bar{c}_p$, $\bar{\gamma}$, or $\bar{K}_a$ and

$$\begin{gathered}\frac{\Delta\gamma}{\gamma_0} = \frac{\Delta c_p}{c_{p0}} - \frac{\Delta c_v}{c_{v0}} = \Big(1 - \frac{1}{\gamma_0}\Big)\Big(\frac{\Delta K}{K_0} + 2\frac{\Delta\alpha}{\alpha_0} - \frac{\Delta c_v}{c_{v0}}\Big),\\ \frac{\Delta K_a}{K_{a0}} = \frac{\Delta K}{K_0} + \frac{\Delta\gamma}{\gamma_0}.\end{gathered} \tag{21}$$

We discuss some particular choices of dispersion laws and see what they mean from the point of view of relaxation theory.

One possible assumption is that there is no dispersion for the quantities $\bar{\alpha}$ and $\bar{c}_p$, i.e., $\Delta\alpha = 0$, $\Delta c_p = 0$. According to (21) or (16) and (17) we then have

$$\frac{\Delta c_v}{c_{v0}} = -\frac{\Delta\gamma}{\gamma_0} = -(\gamma_0 - 1)\frac{\Delta K}{K_0}, \quad \frac{\Delta K_a}{K_{a0}} = \gamma_0\frac{\Delta K}{K_0}. \tag{22}$$

This was just the assumption made in [3] although it corresponds to a rather particular medium. Indeed, when there is a single relaxation time $\tau'(n = 1)$ it follows from (9) that by virtue of $\Delta\alpha = 0$ and $\Delta K \neq 0$ the derivatives with respect to the parameter ξ of the energy and pressure are connected by the relation

$$\rho_0 E_\xi + \alpha_0 T_0 P_\xi = 0.$$

Another particular case corresponds to the Kneser model in which, as we mentioned, $P_\xi = 0$. In accordance with (9) this means that $\Delta\alpha = 0$ and $\Delta K = 0$. The condition (17) is satisfied identically and it follows from (16) and (21) that

$$\Delta c_p = \Delta c_v, \quad \frac{\Delta K_a}{K_{a0}} = \frac{\Delta\gamma}{\gamma_0} = -\Big(1 - \frac{1}{\gamma_0}\Big)\frac{\Delta c_v}{c_{v0}}. \tag{23}$$

A third model was developed on the basis of the theory of Rayleigh scattering developed by Mountain [6] for a liquid. To evaluate the $\omega\mathbf{q}$ intensity of the density fluctuations in the liquid he starts from the hydrodynamics and heat transfer equations (Eqs. (11)–(13) in [6]) which after eliminating the density fluctuations ρ_1 and replacing the velocity $\mathbf{v}$ by grad φ can be written as follows

$$\begin{gathered}\omega^2\rho_0\varphi + \Big(K_0 + \frac{\Delta K i\omega\tau'}{1 + i\omega\tau'} + i\omega\eta_v + \frac{4}{3}i\omega\eta_s\Big)\nabla^2\varphi - \alpha_0 K_0 i\omega T_1 = 0,\\ \alpha_0 K_0 T_0\nabla^2\varphi + \rho_0 c_{v0} i\omega T_1 - \varkappa\nabla^2 T_1 = 0.\end{gathered} \tag{24}$$

Here η_V and η_S are the frequency-independent bulk and shear viscosities introduced by Mountain.[3)] Comparing Eqs. (24) with the general Eqs. (13) shows that

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[3)]In accordance with the Kramers-Kronig relations one must understand the occurrence of frequency-independent viscosities in the sense that in actual fact there is for the compression modulus $\bar{K}$ at least (apart from τ') a second relaxation time $\tau'' \ll \tau'$, but neither τ'' nor the relaxation time τ of the shear modulus occur explicitly if one agrees not to go outside the range of frequencies for which $\omega\tau'' \ll 1$ and $\omega\tau \ll 1$.

one assumes in (24) the product $\overline{\alpha}\overline{K}$ and the specific heat $\overline{c}_V$ to be dispersionless, i.e., $\Delta(\alpha K) = 0$ and $\Delta c_V = 0$. By virtue of (9) this means that in the given case when as before only one relaxation time τ' $(n = 1)$ occurs, the internal energy of the medium is independent of the parameter ξ: $E_\xi = 0$. Equations (24) thus correspond to yet another model of the medium assuming condition (11) but different from the two preceding models. This conclusion is not connected with the requirement of weak dispersion, but if the medium is a low-viscosity one, this model leads in first order in the dispersion drops according to (16) and (21) to the relations

$$\frac{\Delta c_p}{c_{p0}} = \frac{\Delta\gamma}{\gamma_0} = -\left(1 - \frac{1}{\gamma_0}\right)\frac{\Delta K}{K_0}, \quad \frac{\Delta K_a}{K_{a0}} = \frac{\Delta K}{\gamma_0 K_0}. \tag{25}$$

We note, however, at once that the final result of[6], i.e., the formula for the spectral intensity of the scattering does not correspond to the model and viscosity we just described for which all quantities referring to compression ($\overline{K}$, $\overline{\alpha}$, $\overline{c}_V$, $\overline{c}_p$, $\overline{\gamma}$, $\overline{K}_a$) are real while only the shear modulus $\overline{\mu}$ has dispersion. As will be shown below the additional condition that $\overline{K}$ and $\overline{\alpha}$ are real separately, which are not contained in Eqs. (24) appeared in connection with an error let through in[6,7] when the density fluctuations correlation function and thereby the expression for the spectral intensity of the scattered light were derived.

4. ISOTROPIC SCATTERING

The expression for the spectral intensity of isotropic scattering (Eq. (4.1)) obtained in[2] takes the form

$$J_{\mathrm{rad}}(\omega) = (2\pi)^3 |Y|^2 \overline{|u(\omega, \mathbf{q})|^2}$$
$$= \frac{\Theta |Y|^2}{2\pi z}\left\{\frac{\Delta_1(z)}{\Delta(z)} - \frac{\Delta_1(-z)}{\Delta(-z)}\right\} \qquad (z = i\omega). \tag{26}$$

if we neglect the temperature dependence of the dielectric constant ϵ, i.e., limit ourselves to scattering by density fluctuations only. In (26) $\Theta = k_B T_0$ is the temperature of the medium in energy units, ω the shift in frequency reckoned from the frequency of the initial light, $\mathbf{q}$ the scattering vector ($q = 2k\sin(\theta/2)$, where k is the optical wave number in the medium and θ the scattering angle), $Y(i\omega)$ a coefficient which in the case where it is frequency-independent has the value

$$Y_0 = -\rho_0(\partial\varepsilon/\partial\rho)_T,$$

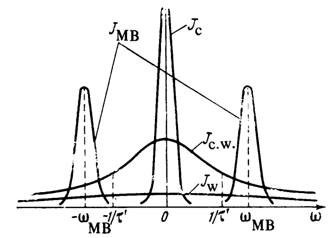
and $\overline{|u|^2}$ is the $\omega\mathbf{q}$ transform of the correlation function of the relative dilatation, i.e., the trace of the deformation tensor $u = u_{\alpha\alpha} = -\rho_1/\rho_0$. The quantities Δ_1 and Δ are in the notation of the present paper given by the equations

$$\Delta_1(z) = (z^2 + \overline{m})\left(\frac{\overline{c}_v}{c_{v0}} + \frac{aq^2}{z}\right) + \frac{T_0}{c_{v0}q^2}|\overline{\alpha}\overline{k}|^2,$$
$$\Delta(z) = \overline{K}(-z)\left[(z^2 + \overline{k} + \overline{m})\left(\frac{\overline{c}_v}{c_{v0}} + \frac{aq^2}{z}\right) + \frac{T_0}{c_{v0}q^2}\overline{\alpha}^2\overline{k}^2\right], \tag{27}$$

where for simplification we have introduced the notation

$$\overline{k} = \overline{K}\frac{q^2}{\rho_0}, \quad \overline{m} = \frac{4}{3}\overline{\mu}\frac{q^2}{\rho_0}, \quad a = \frac{\varkappa}{\rho_0 c_{v0}}.$$

Equations (26) and (27) give (when $\partial\epsilon/\partial T = 0$) the spectral distribution of the intensity of the scattered light for any allowable dispersion laws for the parameters of the medium, simply by substituting in them these dispersion laws. For instance, if there are two relaxation times, τ' for the compression and τ for the shear, it is sufficient to introduce into (26), (27) Eqs. (18) to (20). We shall, however, not give the rather complicated resulting expression but restrict ourselves to the above-mentioned models (22) and (23) of the medium.



The structure of the spectrum is, of course, in general the same both for the two models and for the general case. It consists of the undisplaced line, the Mandel'shtam-Brillouin doublet and two "wings" (see figure) namely, a "shear wing" (background caused by the dispersion in $\overline{\mu}$ and/or the relaxation of the anisotropy, usually simply called the wing) and the "compression wing" (background connected with the dispersion of the quantities $\overline{K}$, $\overline{\alpha}$, and $\overline{c}_V$). In ref.[3] the existence of a "compression wing" in isotropic scattering was first mentioned; its integral intensity was also evaluated there (under condition (22). Mountain[6] also had this wing in mind when he stated the problem of the contribution to the scattering spectrum of "additional" or, as other authors[8] have called them "non-propagating" modes of motion of the viscosity.

The form of the two wings depends essentially on the dispersion laws, in particular, on the magnitude of the relaxation times. In the simplest case of one relaxation time for each kind of wave it is given by the factor $(1 + \omega^2\tau'^2)^{-1}$, and by the same factor but with the time τ for the "shear wing" i.e., the "compression wing" stretches up to frequencies $|\omega| \sim 1/\tau'$, and the "shear wing" occupies a band $|\omega| \lesssim 1/\tau$. As far as the doublet is concerned, when $\tau \ll \tau'$ the position of its lines (shifted ω_{MB} from the undisplaced line) and their width Γ are determined by the dispersion of the "adiabatic" modulus $\overline{K}_a$ (Γ depends also on the thermal conductivity) apart from its dependence (in first approximation) on the form of the connection between $\Delta K_a/K_{a0}$ and $\Delta c_V/c_{V0}$:

$$\omega_{\mathrm{MB}} = \omega_0\left[1 + \frac{q^2\tau'^2\Delta K_a}{2\rho_0(1 + \omega_0^2\tau'^2)}\right],$$

$$\Gamma = \frac{q^2\tau'\Delta K_a}{2\rho_0(1 + \omega_0^2\tau'^2)} + \left(1 - \frac{1}{\gamma_0}\right)\frac{aq^4K_{a0}}{2\rho_0\omega_0^2},$$

where

$$\omega_0^2 = \left(K_{a0} + \frac{4}{3}\mu_0\right)\frac{q^2}{\rho_0} = \gamma_0 k_0 + m_0.$$

The choice of a model of the medium affects, of course, both the total integral intensity of the isotropic scattering and the integral intensities of its separate components. To illustrate this we give without derivation the integral intensities[4)] of the undisplaced line (I_c), the doublet (I_{MB}), and the "compression wing" ($I_{c.w.}$) in the cases (22) and (23), restricting ourselves as before to the first order in the dispersion drops and for both models expressing these corrections in terms of ΔK_a. For the sake of simplicity we take the viscosity to be zero ($\mu_0 = 0$ and hence $\Delta\mu = \mu_\infty = \eta_0/\tau$, $\omega_0^2 = \gamma_0 k_0$) and we shall assume that we can neglect the dispersion of the coefficient Y.

If (22) is valid,[5)]

$$\begin{aligned} I_c &= \frac{\Theta Y_0^2}{K_{a0}}(\gamma_0 - 1)\left(1 - \frac{\Delta K_a}{K_{a0}}\right), \\ I_{MB} &= \frac{\Theta Y_0^2}{K_{a0}}\left\{1 + \frac{\Delta K_a}{K_{a0}}\left[\gamma_0 - 1 - \left(\frac{\omega_0^2\tau'^2}{1+\omega_0^2\tau'^2}\right)^2\right]\right\} - I_w, \\ I_{c.w.} &= \frac{\Theta Y_0^2}{K_{a0}}\frac{\Delta K_a}{K_{a0}}\frac{1}{(1+\omega_0^2\tau'^2)^2}, \\ I_{rad} &= I_c + I_{MB} + I_{c.w.} + I_w \\ &= \frac{\Theta Y_0^2}{K_0}\left[1 + \frac{\Delta K_a}{\gamma_0 K_{a0}}\left(\frac{1-\omega_0^2\tau'^2}{1+\omega_0^2\tau'^2}\right)\right] \end{aligned} \tag{28}$$

while for the Kneser model, i.e., when (23) is valid,

$$\begin{aligned} I_c &= \frac{\Theta Y_0^2}{K_{a0}}(\gamma_0 - 1)\left(1 - \frac{\gamma_0}{\gamma_0 - 1}\frac{\Delta K_a}{K_{a0}}\right), \\ I_{MB} &= \frac{\Theta Y_0^2}{K_{a0}}\left\{1 + \frac{\Delta K_a}{K_{a0}}\left[\gamma_0 - \left(\frac{\omega_0^2\tau'^2}{1+\omega_0^2\tau'^2}\right)^2\right]\right\} - I_w, \\ I_{c.w.} &= \frac{\Theta Y_0^2}{K_{a0}}\frac{\Delta K_a}{K_{a0}}\left(\frac{\omega_0^2\tau'^2}{1+\omega_0^2\tau'^2}\right)^2, \\ I_{rad} &= I_c + I_{MB} + I_{c.w.} + I_w = \frac{\Theta Y_0^2}{K_0}. \end{aligned} \tag{29}$$

Here I_w is the correction for the dispersion of the shear modulus which is equal to the integral intensity of the shear wing:

$$I_w = \Theta Y_0^2 \frac{4q^4\tau^4\Delta\mu}{3\rho_0^2(1+\omega_0^2\tau^2)^2}. \tag{30}$$

An appreciable difference is present between Eqs. (28) and (29) with regard to the dependence of the dispersion corrections both on $\omega_0\tau'$ and on $\gamma_0 = c_{p0}/c_{v0}$.

We do not give the analogous formulae for the third model corresponding to Eq. (25) since in the next section we analyze Mountain's result in the form in which it was obtained in[6], i.e., without restriction to the case of weak dispersion and taking into account frequency-independent viscosities.

5. REMARKS ON MOUNTAIN'S THEORY[6,7]

It was established in Sec. 3 that it is assumed in the initial equations of Mountain's that $\overline{\alpha k} = \alpha_0 k_0$, $\bar{c}_v = c_{v0}$ and, hence,

$$|\overline{\alpha k}|^2 = \bar{\alpha}^2\bar{k}^2 = \alpha_0^2 k_0^2 = k_0(\gamma_0 - 1)c_{v0}q^2/T_0. \tag{31}$$

The last expression follows from the thermodynamic relation

$$c_{p0} = c_{v0} + \frac{T_0}{\rho_0}\alpha_0^2 K_0 = c_{v0} + \frac{T_0}{q^2}\alpha_0^2 k_0,$$

into which the first Eq. (3) goes over when $\omega = 0$.

Substituting (31) into (27) and the result into (26), we get

$$J_{rad}(\omega) = \frac{\Theta Y_0^2}{2\pi z}\left\{\frac{(z^2+\bar{m})\left(1+\frac{aq^2}{z}\right) + k_0(\gamma_0-1)}{\bar{K}(-z)[(z^2+\bar{k}+\bar{m})\left(1+\frac{aq^2}{z}\right)+k_0(\gamma_0-1)]} - \text{c.c.}\right\} \tag{32}$$

(c.c.—complex conjugate quantity). If we follow the scheme of[6] and assume that the frequency dependence of the coefficient of $\nabla^2\varphi$ in (24) is determined by the dispersions of both elastic moduli, i.e.,

$$\bar{k} = \bar{K}\frac{q^2}{\rho_0} = k_0 + q^2 z\left(b_v + \frac{b'}{1+z\tau'}\right), \quad \bar{m} = \frac{4}{3}\bar{\mu}\frac{q^2}{\rho_0} = q^2 z b_s, \tag{33}$$

where $b_v = \eta_v/\rho_0$, $b_s = 4\eta_s/3\rho_0$, and $b' = (\tau'/\rho_0)\Delta K$, and substitute this expression into (32), the result (see below, Eq. (39)) differs from that obtained by Mountain.

If we assume that not only the product $\overline{\alpha k}$, but also α and $\bar{k}$ themselves separately are real, the only frequency dependent parameter turns out to be the shear modulus $\bar{\mu}$, i.e., we must put in the coefficient of $\nabla^2\varphi$

$$\bar{k} = k_0, \quad \bar{m} = q^2 z\left(b_0 + \frac{b'}{1+z\tau'}\right), \tag{34}$$

where

$$b_0 = b_v + b_s = (\eta_v + {}^4/_3\eta_s)/\rho_0.$$

For real $\bar{k}$ Eq. (32) takes the form

$$J_{rad}(\omega) = \frac{\Theta Y_0^2}{2\pi K_0}\left\{\frac{(z^2+\bar{m})(1+aq^2/z) + k_0(\gamma_0-1)}{z[(z^2+k_0+\bar{m})(1+aq^2/z)+k_0(\gamma_0-1)]} + \text{c.c.}\right\}. \tag{35}$$

Substituting here Eq. (34) for $\bar{m}$ and bearing in mind that $k_0\gamma_0 = K_0\gamma_0 q^2/\rho_0 = V_0^2 q^2$ and $z = i\omega$, we are then led to Mountain's result (Eqs. (25) to (27) in[6]), vis.,

$$J_{rad}(\omega) = \frac{\Theta Y_0^2}{\pi K_0}\frac{N_1 D_1 + N_2 D_2}{D_1^2 + D_2^2}, \tag{36}$$

$$N_1 = -\omega^2 + b_0 a q^4 + V_0^2 q^2\left(1 - \frac{1}{\gamma_0}\right) + \frac{b' q^2(\omega^2\tau' + aq^2)}{1+\omega^2\tau'^2}, \tag{37}$$

$$N_2 = \omega q^2\left[a + b_0 + \frac{b'(1 - aq^2\tau')}{1+\omega^2\tau'^2}\right],$$

$$D_1 = q^2\left[-\omega^2(a+b_0) + V_0^2 q^2\frac{a}{\gamma_0} - \frac{b'\omega^2(1-aq^2\tau')}{1+\omega^2\tau'^2}\right], \tag{38}$$

$$D_2 = \omega\left[-\omega^2 + V_0^2 q^2 + b_0 a q^4 + \frac{b' q^2(\omega^2\tau' + aq^2)}{1+\omega^2\tau'^2}\right].$$

These formulae thus reflect the influence on the compression waves exerted by the dispersion of only the shear modulus, to which (in accordance with the initial scheme) are assigned such values of the relaxation time and dispersion drop as in fact the compression modulus possesses. Apparently, we must look just here for an explanation of the satisfactory agreement of the result (36)–(38) with experiment.[8]

We note that Mountain's result which is advantageous because of its simplicity can be even further simplified since the bracket in the denominator in (35) differs from that in the numerator only by terms $k_0(1 + aq^2/z)$. We can thus write (35) in the form

$$J_{rad}(\omega) = -\frac{\Theta Y_0^2}{2\pi K_0}\left\{\frac{k_0(1+aq^2/z)}{z[(z^2+k_0+\bar{m})(1+aq^2/z)+k_0(\gamma_0-1)]} + \text{c.c.}\right\}, \tag{35a}$$

4)The integral intensities are determined by the residues of the integral $\int J_{rad}(z)\,dz$ in the appropriate poles of $J_{rad}(z)$. In a medium with a weak dispersion it is sufficient to find these poles up to first order in the dispersion drops and the thermal conductivity κ. The total intensity I_{rad} can be found at once using the general theorems (6.6)–(6.8) given in[1].

5)Equations (28) correspond to (2.1)–(2.4) in[3] for $\partial\epsilon/\partial T = 0$ and $\mu_0 = 0$ while I_c in (2.1) was given only in zeroth approximation. Unfortunately, there is a misprint in (2.4) and in Eq. (2.2) and (2.9) following from it an error slipped in: in the square bracket the term $\gamma_0 - 1$ was omitted.

whence we get (with the old D_1 and D_2) for N_1 and N_2 instead of (37) the expression

$$N_1 = -k_0 = -\frac{V_0^2 q^2}{\gamma_0}, \quad N_2 = k_0 \frac{aq^2}{\omega} = \frac{V_0^2 aq^4}{\gamma_0 \omega}. \quad (37a)$$

We now give the result of substituting into (32) the dispersion laws (33) in which not the shear modulus μ, but the compression modulus $\overline{K}$ relaxes with relaxation time τ'. For the sake of simplicity we restrict ourselves to the first order in the viscosities b_V and b', and we find instead of (36)

$$J_{\text{из}}(\omega) = \frac{\Theta Y_0^2}{\pi K_0}\left\{\frac{q^2}{k_0}\left(b_v + \frac{b'}{1+\omega^2\tau'^2}\right) + \frac{N_1 D_1 + N_2 D_2}{D_1^2 + D_2^2}\right\}, \quad (39)$$

where D_1 and D_2 are, as before, given by Eqs. (38) and N_1 and N_2 differ from (37a):

$$N_1 = -k_0 + 2aq^4\left(b_v + \frac{b'}{1+\omega^2\tau'^2}\right),$$
$$N_2 = k_0\frac{aq^2}{\omega} - 2q^2\omega\left(b_v + \frac{b'}{1+\omega^2\tau'^2}\right). \quad (40)$$

Of course, as b_V = const the spectral intensity (39) does not vanish as $|\omega| \to \infty$. The evaluation of the integral intensity requires either the dropping of b_V or taking the second relaxation time $\tau'' \ll \tau'$ into account (see Sec. 3). In the admissible range of frequencies $\omega \lesssim 1/\tau'$ the additional terms occurring in (39) and (40) beyond those which are contained in (36) and (37a) affect in turn the total intensity of the "compression wing" without, however, changing its form.

It is naturally of interest to see where in Mountain's derivation the additional condition that $\overline{K}$ (and thereby also $\overline{\alpha}$) be real entered, notwithstanding the author's intentions.

To obtain the autocorrelation function of the density fluctuations ρ_1, the $\omega\mathbf{q}$-transform of which determines $J_{\text{rad}}(\omega)$, Mountain performs on the initial equations for $\rho_1(t, \mathbf{r})$, $\mathbf{v}(t, \mathbf{r})$, and $T_1(t, \mathbf{r})$ a spatial Fourier transformation (and obtained equations for the transforms $\tilde{\rho}_1(t, \mathbf{q}), \ldots$) and a time Laplace transformation (transforms $\hat{\rho}_1(z, \mathbf{q}), \ldots$). It is possible to use the algebraic equations thus obtained to express $\hat{\rho}_1(z, \mathbf{q})$ in terms of the initial ($t = 0$) values of the $t\mathbf{q}$-transforms, i.e., in terms of $\tilde{\rho}_1(0, \mathbf{q})$, $\tilde{\mathbf{v}}(0, \mathbf{q})$, and $\tilde{T}_1(0, \mathbf{q})$. Explicitly this equation is the following:

$$\hat{\rho}_1(z, \mathbf{q}) = \tilde{\rho}_1(0, \mathbf{q})P(z) + i\mathbf{q}\tilde{\mathbf{v}}(0, \mathbf{q})Q(z) + \tilde{T}_1(0, \mathbf{q})R(z), \quad (41)$$

where P, Q, and R are well-defined functions of z, $P(z) = F(z)/G(z)$, where G and F are given by Eqs. (17) and (18) of [6]). The terms with $\tilde{\mathbf{v}}(0, \mathbf{q})$ and $\tilde{T}_1(0, \mathbf{q})$ were, however, dropped by Mountain. The motivation which refers only to $\tilde{T}_1(0, \mathbf{q})$ is given on p. 208 of [7]: "Since the density and temperature are thermodynamically independent it is not necessary to include terms containing $\tilde{T}_1(0, \mathbf{q})$ in the solution for $\hat{\rho}_1(z, \mathbf{q})$."

First of all, it is not clear how one can transfer this argument to the velocity $\tilde{\mathbf{v}}(0, \mathbf{q})$ which for some reason completely dropped out of the argument, although $\mathbf{v}$ is certainly connected with ρ_1 through the equation of continuity.

Secondly, the argument itself is wrong. The thermodynamic independence of equilibrium quantities bears no relation to the problem of the correlation between the thermal fluctuations of these quantities and this is the only essential one for the scattering spectrum.

During the remainder of the derivation, Eq. (41) is multiplied by $\tilde{\rho}_1(0, -\mathbf{q}) = \tilde{\rho}_1^*(0, \mathbf{q})$ and the statistical average is taken. The omissions in [6,7] thus mean that the density fluctuations are assumed to be uncorrelated with the velocity and with the temperature fluctuations. This is correct just when there is no dispersion in the compression modulus, i.e., when $\overline{K} = K_0$.

In [1] we calculated a number of cross $\omega\mathbf{q}$-transforms, amongst whom $\overline{\rho_1 T_1^*}$ (Eq. (5.9) in [1]).[6)] The correlation function in the $t\mathbf{q}$-representation is

$$\overline{\tilde{\rho}_1(t_1, -\mathbf{q})\tilde{T}_1(t_2, \mathbf{q})} = \int_{-\infty}^{+\infty} \overline{\hat{\rho}_1(-\omega, -\mathbf{q})\hat{T}_1(\omega, \mathbf{q})}\, e^{i\omega(t_1 - t_2)}\, d\omega,$$

which for $t_1 = t_2 = 0$ gives

$$\overline{\tilde{\rho}_1(0, -\mathbf{q})\tilde{T}_1(0, \mathbf{q})} = \int_{-\infty}^{+\infty} \overline{\hat{\rho}_1^*(\omega, \mathbf{q})\hat{T}_1(\omega, \mathbf{q})}\, d\omega.$$

This integral and also the analogous integral for $\overline{\mathbf{q}\tilde{\rho}_1(0, -\mathbf{q})\tilde{\mathbf{v}}(0, \mathbf{q})}$ can easily be evaluated using the theorems (6.6) and (6.7) given in [1]. Under the initial restrictions of Mountain's theory ($\overline{\alpha}\overline{K} = \alpha_0 K_0$ and $\overline{c}_V = c_{V0}$) both integrals turn out to be proportional to $\Delta K = K_\infty - K_0$. Hence it follows that dropping the second and third term in (41) means the introduction of the additional condition that $\overline{K}$ be real.

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6) We use this opportunity to correct Eq. (4.14) in [1]: in it one should retain only the first two terms in the braces.