

**THE EFFECT OF SPATIAL DISPERSION ON THE PROPAGATION AND SCATTERING OF WAVES NEAR THE CRITICAL POINT**

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Submitted to JETP editor April 9, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 47, 2134-2149 (December, 1964)

Owing to the large compressibility near the critical state of matter, the nonlocal connection between changes of pressure and density (spatial dispersion) plays an important role. This leads to a dependence of the sound speed on the wavelength and, correspondingly, to a change in the character of the scattering and absorption of both sound and electromagnetic waves. Characteristics of the average and scattered fields (dispersion and absorption of waves, extinction coefficient of light waves, intensity of the Mandel'shtam-Brillouin doublet etc.) are determined on the basis of the general theory of fluctuations of hydrodynamic quantities as extended to the case of media with spatial dispersion. These characteristics can be employed in experimental investigation of the critical transition.

**1. INTRODUCTION**

AS is well known, the curve on which the isothermal sound speed vanishes,

$$(\partial p / \partial \rho)_T = 0, \tag{1.1}$$

separates the thermodynamically stable states of a single-component system from the unstable states; therefore, the thermodynamic potentials can have a singularity on this curve.<sup>[1]</sup> It is evident that only the critical point on this curve, defined, in addition to (1.1) by the condition

$$(\partial^2 p / \partial \rho^2)_T = 0 \tag{1.2}$$

is accessible to experiment.

The other points on the curve (1.1) can be reached only through metastable (superheated or supercooled) states, which clearly excludes the possibility of their experimental investigation.

The critical point of a liquid and a gas determines the termination of the region of two-phase systems; for states above the critical, the difference between the phases disappears. In the existing phenomenological theory,<sup>[1]</sup> the equation of state of the material near the critical point is obtained with the aid of (1.1) and (1.2) as experimental facts. It is here assumed that the thermodynamic potentials have no other singularities at the critical point or, at least, that these singularities do not appear in the approximations customarily used.

Recently, Voronel' et al<sup>[2]</sup> discovered a logarithmic increase in the specific heats  $c_v$  of argon and oxygen as  $T_0 \rightarrow T_c$  ( $\rho_0 = \rho_c$ ); the thermodynamic consequences to which the presence of such a singularity leads were then explained.<sup>1)</sup> Since it follows, as is well known, from (1.1) and (1.2) that in the absence of a singularity,

$$c_p \sim (\partial p / \partial \rho)_T^{-1} \sim (T_0 - T_c)^{-1},$$

$$u_s^2 = (\partial p / \partial \rho)_s \sim 1 / c_v, \tag{1.3}$$

at  $\rho_0 = \rho_c$ , then, owing to the logarithmic singularity of the specific heat, not only the isothermal, but also the adiabatic sound speed  $u_s$  approaches zero:

$$c_v \sim \ln |T_0 - T_c|, \quad u_s^2 \sim 1 / \ln |T_0 - T_c|. \tag{1.4}$$

In this connection, experimental studies of sound propagation and of the scattering of electromagnetic waves close to the critical point are very important for elucidation of the character of the singularities of thermodynamic quantities.

<sup>1)</sup>At the critical point, the heat of transition vanishes, in which connection the transition at the critical point is akin to a second-order transition. For the  $\lambda$  transition in helium, it was discovered experimentally by Fairbank and others<sup>[4]</sup> that there is a logarithmic growth of the heat capacity as  $T \rightarrow T_\lambda$ . Recently, a theoretical analysis by Patashinskiĭ and Pokrovskiĭ<sup>[5]</sup> and Vedenov and Dykhne (private communication) has given evidence of the logarithmic singularity for the  $\lambda$  transition at the critical point as well.

## 2. SPATIAL DISPERSION, ITS EFFECT ON FLUCTUATIONS AND THE SOUND SPEED

A. Out of all the characteristic peculiarities of sound waves near the critical point, of most interest to us is the presence of spatial dispersion, that is, the dependence of the sound speed and the absorption on the wavelength  $2\pi/k$ .<sup>2)</sup>

We shall assume that the nonlinearity, whose role naturally increases as  $u_S \rightarrow 0$ , remains unimportant at sufficiently low intensities of the sound. Actually, the dissipative terms in the hydrodynamic equations have a nondimensional order  $\epsilon_d \sim \nu k^2/\omega \sim \nu\omega/u_S^2$  and as  $u_S \rightarrow 0$  they grow like  $1/u_S^2$  ( $\nu$  is the viscosity; in view of the growth of the specific heat, the term with the thermal conductivity is omitted in the estimate). The nonlinear terms, which have the order  $\epsilon_n \sim kv/\omega \sim v/u_S$  ( $v$  is the amplitude of the velocity in the sound wave) increase like  $1/u_S$ . Therefore, we shall not consider the nonlinearity, assuming that the condition  $\epsilon_n \ll \epsilon_d$  or  $v \ll \nu\omega/u_S$  is satisfied, i.e., it is assumed, in particular that no significant decrease takes place in the viscosity at the critical point. We note that, as was shown by Soluyan and Khokhlov,<sup>[7]</sup> shock wave formation does not take place even for  $\epsilon_n \lesssim \epsilon_d$ .<sup>3)</sup>

The unlimited growth of the compressibility of matter  $\rho^{-1}(\partial\rho/\partial p)_T$  upon approach to the critical point leads to a corresponding increase in density fluctuations, and also in the distance at which the correlation fluctuations become important. The correlation radius, which characterizes these distances, increases sharply close to  $T_C$ , in connection with which the thermodynamic quantities at the different points of the medium are shown to be essentially connected with one another. This is shown in the fact that the equation of state becomes nonlocal close to the critical point. A substance close to  $T_C$  behaves as a medium with spatial dispersion.

In accord with the well known formulas,<sup>[1]</sup> without account of spatial dispersion,

$$\overline{(\Delta\rho)^2} \sim (\partial\rho/\partial p)_T, \quad \overline{(\Delta T)^2} \sim 1/c_v,$$

i.e., as  $T_0 \rightarrow T_C$ , we have  $\overline{(\Delta\rho)^2} \rightarrow \infty$ , and as a con-

sequence of the logarithmic singularity we get  $\overline{(\Delta T)^2} \rightarrow 0$ . The finite expressions for  $(\Delta\rho)^2$  are obtained under the assumption that there is a weak spatial dispersion. This is made clear in the fact that in the nonlinear part of the free energy of an isothermal system  $\Delta F_p = \int dr F(\mathbf{r})$  are considered as quadratic terms both in the deviations of the density  $\rho(\mathbf{r})$  from the equilibrium  $\rho_0$ , and in the density gradients<sup>[1]</sup>

$$F(\mathbf{r}) = \frac{1}{2} a \rho^2 + \frac{1}{2} b (\nabla\rho)^2, \quad \rho(\mathbf{r}) = \tilde{\rho}(\mathbf{r}) - \rho_0, \\ a = \rho^{-1}(\partial p/\partial\rho)_T. \quad (2.1)$$

The values of the fluctuations, correlation functions, intensity of light scattering, etc., are calculated with the help of (2.1).

B. In what follows, we abandon the assumption of weak spatial dispersion, and assume that (as a consequence of the neglect of nonlinear effects discussed above) there is an arbitrary linear nonlocal connection between  $F(\mathbf{r})$  and  $\rho^2$

$$F(\mathbf{r}) = \int K(\mathbf{r}-\mathbf{r}', \mathbf{r}-\mathbf{r}'') \rho(\mathbf{r}') \rho(\mathbf{r}'') d\mathbf{r}' d\mathbf{r}''. \quad (2.2)$$

The medium is assumed to be homogeneous and isotropic, i.e., the gravitational effect is in particular neglected.

For the change in the total free energy we have

$$\Delta F_p = \int Q(\mathbf{r}'-\mathbf{r}'') \rho(\mathbf{r}') \rho(\mathbf{r}'') d\mathbf{r}' d\mathbf{r}'', \\ Q(\mathbf{r}'-\mathbf{r}'') = 2 \int K(\mathbf{r}-\mathbf{r}', \mathbf{r}-\mathbf{r}'') d\mathbf{r}. \quad (2.3)$$

The kernel  $Q(\mathbf{r})$  plays a significant role in the following consideration, defining both the fluctuation and acoustic properties of the system. The general properties of the kernel  $Q$  are discussed below. Here we note only that  $Q(\mathbf{r})$  depends on the temperature and the equilibrium density as parameters, while the dependence on  $\mathbf{r}$  can change materially as  $T_0 \rightarrow T_C$  and  $\rho_0 \rightarrow \rho_C$ . Of most interest obviously is a knowledge of the function  $Q(\mathbf{r}, T_0 - T_C, \rho_C)$ . In what follows, we shall omit the thermodynamic arguments in  $Q$ .

Equation (2.3) is rewritten in Fourier components in the form

$$\Delta F_p = \frac{1}{2V} \sum_{\mathbf{k}} Q(\mathbf{k}) |\rho_{\mathbf{k}}|^2, \quad Q(\mathbf{k}) = Q(-\mathbf{k}) = 2K(\mathbf{k}, -\mathbf{k}). \quad (2.4)$$

The Fourier components are defined by the relations

$$\rho(\mathbf{r}) = \frac{1}{V} \sum_{\mathbf{k}} \rho_{\mathbf{k}} e^{i\mathbf{k}\mathbf{r}}, \quad \rho_{\mathbf{k}} = \int \rho(\mathbf{r}) e^{-i\mathbf{k}\mathbf{r}} d\mathbf{r}, \quad (2.5)$$

while, by assuming  $V$  to be sufficiently large, we also replace the sum over  $\mathbf{k}$  by an integral:

<sup>2)</sup>We limit ourselves to the single-phase region around the critical point. In the region of stratification of phases, additional mechanisms of dispersion and nonlinearity must operate.<sup>[6]</sup>

<sup>3)</sup>The conditions for shock-wave formation in a medium with spatial dispersion have not been studied as yet. However, it is natural to expect that spatial dispersion must make the formation of discontinuities more difficult.

$$\rho(\mathbf{r}) = \int \frac{d\mathbf{k}}{(2\pi)^3} \rho_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} \quad (2.6)$$

In accord with the Boltzmann principle, it follows from (2.4) for the density fluctuations in the volume  $V$  ( $k_B$  is the Boltzmann constant) that

$$|\overline{\rho_{\mathbf{k}}}|^2 = V k_B T / Q(\mathbf{k}). \quad (2.7)$$

The weak spatial dispersion corresponds to the following form of the kernel  $K$ :

$$K(\mathbf{r} - \mathbf{r}', \mathbf{r} - \mathbf{r}'') = \frac{1}{2} a \delta(\mathbf{r} - \mathbf{r}') \delta(\mathbf{r} - \mathbf{r}'') + \frac{1}{2} b \nabla_{\mathbf{r}'} \delta(\mathbf{r} - \mathbf{r}') \nabla_{\mathbf{r}''} (\mathbf{r} - \mathbf{r}''). \quad (2.8)$$

Here (2.3) transforms into (2.1).

From (2.3) and (2.4) we find in this case

$$Q(\mathbf{k}) = a + b k^2, \quad (2.9)$$

which, after substitution in (2.7), leads to the well known formula of Ornstein and Zernike for the fluctuation of the density near the critical point.

The relation (2.9) is valid only in the case of sufficiently small correlation radii  $r_0$ . According to<sup>[1]</sup> the correlation radius  $r_0 = (b/a)^{1/2} \rightarrow \infty$  like  $(T_0 - T_C)^{-1/2}$  (cf. (2.1) and (1.3)), and it must be expected that Eq. (2.9) ceases to be valid in the immediate vicinity of  $T_C$ . In this very case, however, if the thermodynamic potentials have a singularity at  $T_C$ , the expansions (2.8) and (2.9) generally do not hold.

In the general case, the form of  $Q(\mathbf{k})$  cannot be obtained in phenomenological theory and must be determined by comparison with experiment. As such experiments, in addition to acoustic ones, one can use the phenomenon of critical opalescence, since the intensity of the scattered light is determined by the values of (2.7), where  $\mathbf{k}$  is the change in the wave vector of the light in the scattering process.<sup>[8]</sup>

When considering wave scattering it is more convenient to use an average characteristic of the fluctuations, which does not depend on the volume. In this case, we introduce the correlation function  $W^\rho(\mathbf{r} - \mathbf{r}')$ :

$$\overline{\rho(\mathbf{r}) \rho(\mathbf{r}') } = \overline{\rho^2} W^\rho(\mathbf{r} - \mathbf{r}'). \quad (2.10)$$

Comparing the definitions (2.6) and (2.10), we get

$$|\overline{\rho_{\mathbf{k}}}|^2 = \overline{\rho^2} V W^\rho(\mathbf{k}). \quad (2.11)$$

Thus, in accord with (2.7) and (2.11),  $Q(\mathbf{k})$  is directly connected with the Fourier component of the correlation function:

$$[\overline{\rho(\mathbf{r}) \rho(\mathbf{r}')}]_{\mathbf{k}} = \overline{\rho^2} W^\rho(\mathbf{k}) = k_B T / Q(\mathbf{k}). \quad (2.12)$$

C. We shall now show that the function  $\rho_0 Q(\mathbf{k})$

represents the isothermal speed of sound with wavelength  $2\pi/k$ .

The work  $\delta R$  expended on a unit mass when changing the density by a value  $\rho$  is evidently  $\delta R = \rho_0^{-2} (p_0 + p) \rho$ , where  $p(\mathbf{r})$  is the deviation of the pressure from the equilibrium value  $p_0$ . The work performed on the entire body is equal to

$$\Delta R = \int \frac{p(\mathbf{r})}{\rho_0} \rho(\mathbf{r}) d\mathbf{r}. \quad (2.13)$$

For  $T = \text{const}$ , we have  $\Delta R = \Delta F_p$ . Comparing (2.13) with (2.3), we find that there exists a non-local connection between the change in pressure and density:

$$p(\mathbf{r}) = \rho_0 \int Q(\mathbf{r} - \mathbf{r}') \rho(\mathbf{r}') d\mathbf{r}'. \quad (2.14)$$

This relation becomes evident also when we use functional derivatives to replace the ordinary derivatives in the well known thermodynamic formulas (for  $T = \text{const}$ ):

$$p_0 + p(\mathbf{r}) = \rho_0 \frac{\delta F_p}{\delta \rho(\mathbf{r})}, \quad \Delta F_p = \int d\mathbf{r}' \rho(\mathbf{r}') \frac{\delta F_p}{\delta \rho(\mathbf{r}')},$$

$$Q(\mathbf{r} - \mathbf{r}') = \frac{\delta^2 F_p}{\delta \rho(\mathbf{r}) \delta \rho(\mathbf{r}')}. \quad (2.15)$$

In the presence of spatial dispersion, the speed of sound is an operator. The square of the isothermal speed of sound should be determined as the variational derivative of the pressure with respect to the density:

$$u_T^2(\mathbf{r} - \mathbf{r}') \equiv \delta p(\mathbf{r}) / \delta \rho(\mathbf{r}') = \rho_0 Q(\mathbf{r} - \mathbf{r}'). \quad (2.16)$$

The Fourier components of this quantity  $u_T(\mathbf{k})$  have the physical meaning of the sound speed (for a disturbance with a given value of the wave vector  $\mathbf{k}$ ); we get for  $u_T(\mathbf{k})$  from (2.12),

$$u_T^2(\mathbf{k}) = \rho_0 Q(\mathbf{k}) = \rho_0 k_B T / \overline{\rho^2} W^\rho(\mathbf{k}). \quad (2.17)$$

This relation is analogous to the well known quantum relation of Feynman,<sup>[9]</sup> which connects the correlation function with the excitation spectrum in liquid helium.

The adiabatic sound speed, as is not difficult to establish, has a Fourier component  $u_S(\mathbf{k})$  equal to

$$u_S^2(\mathbf{k}) = u_T^2(\mathbf{k}) + \frac{T_0}{c_v \rho_0^2} \left( \frac{\partial p}{\partial T} \right)_\rho^2. \quad (2.18)$$

Here, by virtue of the smallness of the temperature fluctuations, the assumption is used that there is a local connection between the pressure and the temperature, owing to which  $(\partial p / \partial T)_\rho$  does not depend on  $\mathbf{k}$  and has the meaning of the ordinary thermodynamic derivative; that is, for the case  $T \neq \text{const}$ , (2.14) can be written in the form

$$p(\mathbf{r}) = \rho_0 \int Q(\mathbf{r} - \mathbf{r}') \rho(\mathbf{r}') d\mathbf{r}' + \left( \frac{\partial p}{\partial T} \right)_\rho T(\mathbf{r}). \quad (2.19)$$

The dispersion equation for sound waves, with neglect of viscosity and thermal conductivity, has the usual form  $\omega^2 = u_s^2(\mathbf{k})k^2$ , but the sound speed is a function of the wave vector.<sup>4)</sup>

The quantity  $Q(\mathbf{k})$  is real, i.e., the spatial dispersion itself does not lead to sound absorption. Account of the dispersion coefficients requires, generally speaking, account also of temporal dispersion. The relations which generalize (2.12) and (2.17) are obtained below with the help of the general Landau and Lifshitz theory of the fluctuation of hydrodynamic quantities<sup>[10]</sup>, applied to the case of media with spatial dispersion.<sup>5)</sup>

### 3. HYDRODYNAMIC FLUCTUATIONS IN A MEDIUM WITH SPATIAL DISPERSION

A. According to the fluctuation-dissipation theory, the temporal Fourier component of the correlation function of the variable  $x$  can be expressed in terms of an entirely different problem of the response of the system to the influence of an external force  $f$  of the same frequency  $\omega$ , which leads to a deviation of the value of  $x$  from its equilibrium value. It is convenient to unite these two problems formally by considering that the fluctuations of the value of  $x$  are brought about by the action on the system of an external random force  $f$ . From this view point, the content of the fluctuation-dissipation theorem reduces to the result that the power entering the system because of the work of the external forces must be entirely dissipated and given off to the thermostat in order that the equilibrium state of the system not be disturbed.

All the general relations of the theory of fluctuations in the form proposed by Landau and Lifshitz<sup>[8]</sup> and by Rytov<sup>[12]</sup> are directly applicable to the case of spatial dispersion, if one takes it into account that the generalized coordinates  $x_n(\mathbf{r})$  depend not only on discrete ( $n$ ), but also on continuous parameters—the coordinate  $\mathbf{r}$ .

<sup>4)</sup>The logarithmic singularity of the specific heat near the  $\lambda$  point and the critical point (see the footnote 1) is connected with the existence of quasiparticles with occupation numbers  $n_{\mathbf{k}} \sim |\rho_{\mathbf{k}}|^2$  and with dispersion law  $\epsilon_{\mathbf{k}} = Ak^{3/2}$ . Thus, for sufficiently small  $\mathbf{k}$ , one has  $Q(\mathbf{k}) = a + bk^{3/2}$ . Here the correlation function for  $T_0 = T_c$  has the form  $W(\mathbf{r}) \sim r^{-3/2}$  (instead of  $W(\mathbf{r}) \sim r^{-1}$  for (2.9)). The dispersion law of sound at the critical point has a form close to parabolic,  $\omega = (\rho_0 B)^{1/2} k^{7/4}$ , where the coefficient  $B$  is equal in order of magnitude to  $B \sim d^{3/2} u^2 / \rho_0 \sim 10^{-2} \frac{\text{cm}^{13/2}}{\text{g sec}^2}$ .

<sup>5)</sup>The extension of the theory of electromagnetic fluctuations to the case of spatial dispersion was made by Bass and Kaganov.<sup>[11]</sup>

The equations which describe the motion of a system under the action of a force  $f$  give the relation between  $x$  and  $f$  at the frequency  $\omega$ . Solving these equations, we find

$$x_n(\mathbf{r}) = \sum_m \int d\mathbf{r}' \alpha_{nm}(\mathbf{r}, \mathbf{r}') f_m(\mathbf{r}'). \quad (3.1)$$

The quantities  $\alpha_{nm}(\mathbf{r}, \mathbf{r}')$  represent the  $\omega$  component of the Green's tensor equations of motion which connect  $f$  and  $x$ . According to the fluctuation-dissipation theorem, the Fourier components of the correlation functions can be expressed in their terms.<sup>[8]</sup>

In a homogeneous medium, the kinetic coefficients  $\alpha$  are functions of the difference in the coordinates  $\alpha_{nm}(\mathbf{r}, \mathbf{r}') = \alpha_{nm}(\mathbf{r} - \mathbf{r}')$ , in which connection it is convenient to transform to the spatial Fourier components of the correlation function

$$(x_n(\mathbf{r}) x_m(\mathbf{r}'))_{\omega, \mathbf{k}} = \frac{i\hbar}{4\pi} (2\pi)^3 (\alpha_{nm}(\omega, \mathbf{k}) - \alpha_{mn}^*(\omega, \mathbf{k})) \text{cth} \frac{\hbar\omega}{2k_B T}. \quad (3.2)^*$$

The dependence on  $\omega$  written above is omitted here and account is taken of the fact that the  $\mathbf{k}$  component of  $\alpha^*(-\mathbf{r})$  is equal to  $\alpha^*(\mathbf{k})$ .

The generalized relations of the symmetry of the kinetic coefficients (the Onsager relations) take the form

$$\alpha_{nm}(\mathbf{r}, \mathbf{r}') = \pm \alpha_{mn}(\mathbf{r}', \mathbf{r}), \alpha_{nm}(\omega, \mathbf{k}) = \pm \alpha_{mn}(\omega, -\mathbf{k}). \quad (3.3)$$

The symbols  $\pm$  in (3.3) refer to the cases in which the generalized coordinates  $x_n$  and  $x_m$  in (3.2) have respectively the same or different symmetry relative to time reversal.

The Fourier components of the correlation function  $(x_n x_m)_{\omega, \mathbf{k}} = x^2 W_{nm}(\omega, \mathbf{k})$  are connected with the correlators of the components of the Fourier quantities (in a homogeneous, stationary system) by the following relation<sup>[8]</sup> (cf. (2.11)):

$$\overline{x_n(\omega, \mathbf{k}) x_m(\omega', \mathbf{k}')} = (x_n x_m)_{\omega, \mathbf{k}} \delta(\omega + \omega') \delta(\mathbf{k} + \mathbf{k}'). \quad (3.4)$$

B. The rate of change of the energy of the liquid is<sup>[6]</sup>

$$\frac{dU}{dt} = \int \frac{\sigma'_{ik}}{2} \left( \frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i} \right) dV - \int q_i \frac{\nabla_i T}{T_0} dV + \int \frac{p}{\rho_0} \frac{\partial \rho}{dt} dV. \quad (3.5)$$

The last term follows from the component  $-pd(1/\rho)$  in the thermodynamic identity for the energy, and is real in the case of spatial dispersion.

Equation (3.5) takes a more convenient form

\*cth = coth.

in terms of the total stress tensor  $\sigma_{ik} = \sigma_{ik}' - p\delta_{ik}$ , with account of the equation of continuity:

$$\frac{dU}{dt} = \int \frac{\sigma_{ih}}{2} \left( \frac{\partial v_i}{\partial x_h} + \frac{\partial v_h}{\partial x_i} \right) dV - \int q_i \frac{\nabla_i T}{T_0} dV. \quad (3.6)$$

As a generalization of (2.19) we assume non-local coupling between the temporal Fourier components of pressure and density:

$$p(\mathbf{r}, \omega) = \int d\mathbf{r}' \rho_0 Q(\mathbf{r} - \mathbf{r}', \omega) \rho(\mathbf{r}', \omega) + \left( \frac{\partial p}{\partial T} \right)_\rho T(\mathbf{r}, \omega), \quad (3.7)$$

and express the values of  $\sigma_{ik}'$  and  $\mathbf{q}$  in the usual fashion in terms of the gradients of the velocities and the temperatures, that is, in essence, spatial dispersion of only the second viscosity is taken into account.

As is well known,<sup>[8]</sup> the correspondence between the quantities  $\mathbf{f}$  and  $\mathbf{x}$  is established with the aid of the equation

$$dU/dt = - \sum_n \int d\mathbf{r} f_n(\mathbf{r}) x_n(\mathbf{r}). \quad (3.7a)$$

Comparison with (3.6) makes it possible to determine two types of generalized forces and coordinates, which differ in the number of indices in the corresponding quantities:<sup>6)</sup>

$$\begin{aligned} f_{nm} &= \frac{\sigma_{nm}^{\text{ext}}}{i\omega}, & x_{nm} &= \frac{i}{2} (k_n v_m + k_m v_n), \\ f_i &= \frac{-q_i^{\text{ext}}}{i\omega}, & x_i &= \frac{ik_i T}{T_0}. \end{aligned} \quad (3.8)$$

Here and throughout this section, we shall, without speaking of it each time, use the Fourier components of the quantities  $\rho$ ,  $T$ ,  $\mathbf{v} \sim e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}$ .

For the calculation of the correlators of hydrodynamic quantities of the type (3.2), it is necessary to find the Green tensor of the hydrodynamic equa-

<sup>6)</sup>The correlators for the extraneous flows  $\sigma_{ik}^{\text{ext}}$  and  $\mathbf{q}^{\text{ext}}$ , with account of spatial dispersion, are obtained from the formulas of Landau and Lifshitz [<sup>9</sup>] by adding to  $\text{Re}\zeta(\omega)\delta(\mathbf{r}-\mathbf{r}')$  the component  $(\rho_0^2/\omega)\text{Im}Q(\mathbf{r}-\mathbf{r}', \omega)$ . Inasmuch as the spatial dispersion is essentially connected with infinite compressibility, then it is natural to assume that it will in no way be significant in the first viscosity and in thermal conductivity, although it is difficult to write down the general relations. For this purpose, it would be necessary to replace in [<sup>9</sup>]

$$\eta(\omega)\delta(\mathbf{r}-\mathbf{r}'), \quad \zeta(\omega)\delta(\mathbf{r}-\mathbf{r}') \quad \text{and} \quad \varkappa(\omega)\delta(\mathbf{r}-\mathbf{r}')$$

by the corresponding relations

$$\eta(\mathbf{r}-\mathbf{r}', \omega), \quad \zeta(\mathbf{r}-\mathbf{r}', \omega), \quad \varkappa(\mathbf{r}-\mathbf{r}', \omega),$$

the definition of which is evident. In our case, we kept  $\zeta(\omega)$ , along with  $Q(\mathbf{r}-\mathbf{r}', \omega)$ , only in order that the formulas have the customary form with the addition of an extra term with dispersion.

tions. The set of linear equations with extraneous flows

$$\begin{aligned} -i\omega\rho_0 v_i &= -ipk_i - k^2\eta v_i - \left( \zeta + \frac{\eta}{3} \right) (\mathbf{k}\mathbf{v}) k_i + ik_i \sigma_{il}^{\text{ext}}, \\ -i\omega\rho &= -i(\mathbf{k}\mathbf{v})\rho_0, \quad p = \rho_0 Q(\mathbf{k}, \omega) \rho + (\partial p/\partial T)_\rho T, \\ -i\omega\rho_0 T_0 S &= -\varkappa k^2 T - ik\mathbf{q}^{\text{ext}}, \end{aligned} \quad (3.9)$$

has the following solution for an unbounded medium

$$\begin{aligned} v_i &= (\omega/\rho_0 k^2) R J k_i (\mathbf{k}\mathbf{q}^{\text{ext}}) - M_{il} k_m \sigma_{lm}^{\text{ext}}, \\ T &= F(\mathbf{k}\mathbf{q}^{\text{ext}}) - (\omega T_0/\rho_0 k^2) R J k_i k_i \sigma_{il}^{\text{ext}}, \\ \rho &= R J(\mathbf{k}\mathbf{q}^{\text{ext}}) - (J/k^2) k_i k_i \sigma_{il}^{\text{ext}} \end{aligned} \quad (3.10)$$

where the following notation is introduced:

$$\begin{aligned} F &= \left[ \frac{1}{\rho_0 c_v} + \frac{\omega T_0}{c_v \rho_0^2} \left( \frac{\partial p}{\partial T} \right)_\rho R J \right] \left( \omega + i \frac{\varkappa k^2}{\rho_0 c_v} \right), \\ M_{il} &= \left\{ \delta_{il} + \frac{k_i k_l}{k^2} \left[ \rho_0 \bar{Q} - i \frac{\omega}{\rho_0} \left( \zeta + \frac{\eta}{3} \right) \right] J \right\} / (\omega \rho_0 + ik^2 \eta), \\ R &= \left( \frac{\partial p}{\partial T} \right)_\rho / \rho_0 c_v \left( \omega + i \frac{\varkappa k^2}{\rho_0 c_v} \right), \\ J &= k^2 \left[ \omega^2 - k^2 \rho_0 \bar{Q} + i \frac{\omega k^2}{\rho_0} \left( \frac{4}{3} \eta + \zeta \right) \right], \\ \rho_0 \bar{Q} &= \rho_0 Q + \frac{T_0}{\rho_0^2} \frac{(\partial p/\partial T)_\rho^2}{c_v + i\varkappa k^2/\omega \rho_0}. \end{aligned} \quad (3.11)$$

We note that for  $\varkappa = 0$ , the value of  $\rho_0 \bar{Q}$  has the meaning of an adiabatic sound speed (with frequency  $\omega$  and wave vector  $\mathbf{k}$ ). Substituting now in (3.9) the generalized forces and coordinates (3.8), we get relations of the type (3.1), from which we can easily find the values of  $\alpha$ :

$$\begin{aligned} \alpha_{il} &= \omega k_i k_l F / T_0, \quad \alpha_{mn, pr} = \omega k_p (k_m M_{nr} + k_n M_{mr}) / 2, \\ \alpha_{i, mn} &= \omega^2 k_i k_m k_n R J / \rho_0 k^2, \quad \alpha_{lm, i} = \omega^2 k_l k_m k_i R J / \rho_0 k^2. \end{aligned} \quad (3.12)$$

As is seen from (3.12), the Onsager relations (3.3) are satisfied.

The values given for the generalized kinetic coefficient make it possible, according to (3.2), taken in the classical limiting case

$$\text{cth} \frac{\hbar\omega}{2k_B T} \rightarrow \frac{2k_B T}{\hbar\omega} \quad (3.12a)$$

to write down the correlation coefficients for all the hydrodynamic quantities. For the density and temperature, we have

$$\rho = \rho_0 k_i v_i / \omega = \rho_0 x_{mm} / i\omega, \quad T = T_0 k_i x_i / ik^2, \quad (3.12b)$$

that is,

$$(\rho(\mathbf{r})\rho(\mathbf{r}'))_{\omega, \mathbf{k}} = \frac{\rho_0^2}{\omega^2} (x_{mm}(\mathbf{r})x_{mm}(\mathbf{r}'))_{\omega, \mathbf{k}},$$

$$(T(\mathbf{r})T(\mathbf{r}'))_{\omega, \mathbf{k}} = \frac{T_0^2 k_i k_k}{k^4} (x_i(\mathbf{r})x_k(\mathbf{r}'))_{\omega, \mathbf{k}},$$

$$(\rho(\mathbf{r})T(\mathbf{r}'))_{\omega, \mathbf{k}} = \frac{\rho_0 T_0}{\omega k^2} k_i (x_{mm}(\mathbf{r}) x_i(\mathbf{r}'))_{\omega, \mathbf{k}}. \quad (3.13)$$

As an example, we take the expression for the Fourier component of the correlators of density and temperature (see<sup>[12]</sup>):

$$\begin{aligned} (\rho(\mathbf{r})\rho(\mathbf{r}'))_{\omega, \mathbf{k}} &= -\frac{k_B T_0 \rho_0}{\pi \omega} (2\pi)^3 \text{Im} J, \\ (T(\mathbf{r})T(\mathbf{r}'))_{\omega, \mathbf{k}} &= -\frac{k_B T_0^2}{\pi} (2\pi)^3 \text{Im} F, \\ (\rho(\mathbf{r})T(\mathbf{r}'))_{\omega, \mathbf{k}} &= -\frac{k_B T_0^2}{\pi} (2\pi)^3 \text{Im} (RJ). \end{aligned} \quad (3.14)$$

To establish the connection with the considerations given above, we note that the real quantities  $Q(\mathbf{r} - \mathbf{r}', 0)$  and  $Q(\mathbf{k}, 0)$  enter in the formulas of Sec. 2. (The absence of time dispersion means that  $Q(\mathbf{r}, t) = Q(\mathbf{r})\delta(t)$ .) In contrast with these,  $Q(\mathbf{r} - \mathbf{r}', \omega)$  and  $Q(\mathbf{k}, \omega)$  are complex quantities.

Carrying out an inverse (spatial) Fourier transformation in (3.14), we have

$$\begin{aligned} (\rho(\mathbf{r})\rho(\mathbf{r}'))_{\omega} &= -\frac{k_B T_0 \rho_0}{\pi \omega} \\ &\times \text{Im} \int \frac{d\mathbf{k}}{(2\pi)^3} \frac{k^2 e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')}}{\omega^2 - k^2 \rho_0 \tilde{Q} + i\omega k^2 (4/3\eta + \zeta)/\rho_0}. \end{aligned} \quad (3.15)$$

Using the Kramers-Kronig relations, as was done by Landau and Lifshitz,<sup>[8]</sup> we get for the correlations at a given moment of time

$$\overline{(\rho(\mathbf{r}, t)\rho(\mathbf{r}', t))} = k_B T_0 \int \frac{d\mathbf{k}}{(2\pi)^3} \frac{e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')}}{Q(\mathbf{k}, 0)}, \quad (3.16)$$

which agrees with the expression (2.12) obtained above, but (3.16) is obtained under more general assumptions—without neglect of viscosity and thermal conductivity.

#### 4. SOUND PROPAGATION CLOSE TO THE CRITICAL POINT

A. The spectrum of hydrodynamic excitations is determined by the poles of the Green's tensors (3.11) of the hydrodynamic equations (3.9). The poles  $\omega\rho_0 = ik^2\eta$  and  $\omega\rho_0 = -ik^2/c_V$  correspond respectively to vortex and entropy (temperature) "waves." The dispersion equation which contains the acoustic branch has the form

$$\begin{aligned} \left(\frac{\omega}{k}\right)^2 - \rho_0 Q(\mathbf{k}, \omega) - \frac{T_0}{\rho_0^2} \frac{(\partial p/\partial T)_\rho^2}{c_V + i\alpha k^2/\omega\rho_0} \\ + i\frac{\omega}{\rho_0} \left(\frac{4}{3}\eta + \zeta\right) = 0. \end{aligned} \quad (4.1)$$

We now consider some special cases.

1. In the immediate vicinity of the critical point, in accord with (1.4),  $c_V$  increases without limit, and one can expand Eq. (4.1) in the small quantity  $1/c_V$ .

Then, if the viscosity is not too large ( $\eta/\rho_0 < \rho_0 Q/\omega$ ), the acoustic wave is propagated with isothermal speed  $u_T(\mathbf{k}, \omega)$ . The absorption takes place as the result of viscosity, and not by virtue of thermal conductivity, if the kinetic coefficients do not have singularities at the critical point. Thus the conclusions are physically understandable, for owing to the infinite growth of the heat conductivity upon approach to the critical point the propagation of a sound wave in such a medium is practically unaccompanied by a change in temperature.

2. In the critical region, but not in the immediate vicinity of the critical point, when there is still no singularity and the inequality  $(kr_0)^2 \ll 1$  is satisfied ( $r_0$  is the correlation radius), that is, where one can consider the spatial dispersion to be weak,  $Q(\mathbf{k})$  has the form of (2.9). One then gets [from Eq. (4.1)] a small [ $\sim (kr_0)^2$ ] correction to the adiabatic sound velocity in the absence of dispersion.<sup>[13]</sup>

3. The dependence of  $Q$  on  $\omega$ , i.e., the temporal dispersion of the second viscosity considered by Mandel'shtam and Leontovich, is important in the presence of slow processes if the period of the sound wave is comparable with the relaxation time in the system. In the critical region, the macroscopic processes are materially slowed, and one can expect the appearance of long relaxation times. In the simplest case, in which there is a single relaxation time  $\tau$ , the dependence of  $Q$  on  $\omega$  would have the form

$$Q(\omega) = (u_0^2 + u_\infty^2 i\omega\tau) / \rho_0(1 + i\omega\tau).$$

Here, in accord with<sup>[6]</sup>, the coefficient of absorption along the wave is proportional to  $\omega\tau$  (for  $\omega\tau \ll 1$ ) or  $1/\omega\tau$  (for  $\omega\tau \gg 1$ ).

B. The scattering by the fluctuations also leads to the appearance of dispersion and additional sound absorption<sup>[14-17]</sup> due to the pumping of energy into the fluctuating part of the field. In view of the specific nature of the critical point, it suffices to consider scattering only by density fluctuations, which we shall consider to be thermal only at the end of the calculation.

In the system of hydrodynamic equations, the terms containing the viscosity and thermal conductivity will be omitted for simplicity in the calculation of additional absorption.

We now separate the various components of the general density  $\tilde{\rho}(\mathbf{r})$  and velocity  $\tilde{\mathbf{v}}(\mathbf{r})$ :

$$\tilde{\rho} = \rho_0 + n + \rho, \quad n = n_0 + n', \quad \tilde{\mathbf{v}} = \mathbf{v} + \mathbf{v}'. \quad (4.2)$$

Here  $\rho_0$  is the equilibrium density,  $\rho$  the fluctuating change in density,  $n$  the density change connected with the passage of the sound wave, while  $n_0$  is its

regular (average) value and  $n'$  its fluctuating part. Similarly,  $\mathbf{v}$  and  $\mathbf{v}'$  are the regular and fluctuating parts of the velocity in the sound wave. By definition,  $\bar{\rho} = \bar{n}' = \bar{\mathbf{v}}' = 0$ .

We carry out a linearization of the hydrodynamic equations in the variables  $n$  and  $\mathbf{v}$  associated with the passage of the sound wave. However, it is necessary to keep the nonlinear terms which describe the coupling of the sound field  $n$  with the thermal fluctuations of the density  $\rho$ . Here, using the condition (1.2), and neglecting the nonlinear terms which contain  $(\partial^2 p / \partial \rho^2) T_C$ , we get

$$\begin{aligned} \frac{\partial n}{\partial t} + \operatorname{div}(\rho_0 \mathbf{v}) &= -\operatorname{div}(\rho \mathbf{v}), \\ \frac{\partial \mathbf{v}}{\partial t} + \frac{\rho_0}{\rho_0 + \rho} \int d\mathbf{r}' dt' n(\mathbf{r}', t') \nabla_{\mathbf{r}} \tilde{Q}(\mathbf{r} - \mathbf{r}', t - t') \\ &= \frac{\rho_0 n}{(\rho_0 + \rho)^2} \int d\mathbf{r}' dt' \rho(\mathbf{r}', t') \nabla_{\mathbf{r}} \tilde{Q}(\mathbf{r} - \mathbf{r}', t - t'). \end{aligned} \quad (4.3)$$

We shall consider the stationary and nonstationary fluctuations separately.

C. In the stationary case, we assume that  $\rho$  does not depend on  $t$ ; eliminating  $\mathbf{v}(\mathbf{r})$  from the system (4.3), we get an equation for  $n(\mathbf{r}, \omega)$ .

The resultant equation, like the Maxwell equations in a fluctuating medium (see Sec. 5B), is of the form  $L_0 n = l(\rho)n$ , where  $l(\rho)$  is an operator defined with the help of the random function  $\rho(\mathbf{r})$ . An approximate method of solution of such an equation, suggested by I. Lifshitz et al.<sup>[14]</sup>, is given below. Averaging the initial equation and subtracting the resulting equation from the initial one, we get

$$L_0 n_0 = \bar{l} n', \quad L_0 n' = l n_0 + (l n' - \bar{l} n'). \quad (4.4)$$

Limiting ourselves to the first approximation, we neglect the components  $l n' - \bar{l} n'$  in (4.4) in comparison with  $l n_0$ . Account of higher approximations calls as usual for involved calculations.<sup>[17]</sup> From the first equation of (4.4), we find  $n' = L_0^{-1} l n_0$ , where  $L_0^{-1}$  is the Green operator inverse to  $L_0$ . Substituting this value of  $n'$  in the first of Eqs. (4.4), we get the initial equation for the mean field in first approximation:

$$(L_0 - \bar{l} L_0^{-1} l) n_0 = 0. \quad (4.5)$$

The Fourier component of (4.5) determines the dispersion equation.<sup>7)</sup> In our case, it is very com-

plicated because of the presence of the quadruple correlator

$$\langle \rho(\mathbf{r}''') \rho(\mathbf{r}'') / [\rho_0 + \rho(\mathbf{r}')] [\rho_0 + \rho(\mathbf{r})] \rangle$$

of the random function  $\rho(\mathbf{r})$ . We limit ourselves to consideration of small fluctuations in the density of a medium with an arbitrary dispersion and large fluctuations in a medium with small spatial dispersion.

1. For small fluctuations, we can expand  $(\rho_0 + \rho)^{-1}$  in a series and limit ourselves to the first term in the expansion. We then get from (4.5) the dispersion equation

$$\begin{aligned} q^2 \rho_0 \tilde{Q}(\mathbf{q}) - \omega^2 - \frac{\bar{\rho}^2}{\rho_0^2} \int \frac{d\mathbf{p}}{(2\pi)^3} \frac{\rho_0^2 \tilde{Q}^2(\mathbf{q} - \mathbf{p}) W^\rho(\mathbf{q} - \mathbf{p})}{\rho_0 \tilde{Q}(\mathbf{p}) p^2 - \omega^2} \\ \times [p^2 q^2 + (\mathbf{p}\mathbf{q})^2] = 0. \end{aligned} \quad (4.6)$$

In (4.6) the Fourier component of the correlation function of the density fluctuations  $W^\rho(\mathbf{q} - \mathbf{p})$  defined in (2.10) is introduced; here (4.6) is valid for sound scattering by density fluctuations of arbitrary origin, for example, of turbulent origin. For thermal density fluctuations, the relation (2.12) is valid and (4.6) takes the form

$$q^2 \rho_0 \tilde{Q}(\mathbf{q}) - \omega^2 - k_B T \int \frac{d\mathbf{p}}{(2\pi)^3} \frac{\tilde{Q}(\mathbf{q} - \mathbf{p}) [p^2 q^2 + (\mathbf{p}\mathbf{q})^2]}{\rho_0 \tilde{Q}(\mathbf{p}) p^2 - \omega^2} = 0. \quad (4.7)$$

Dispersion of the sound velocity and additional absorption can be calculated from (4.7) for an arbitrary value of the spatial dispersion. Thus, for  $\tilde{Q}(\mathbf{k}) = Bk^{3/2}$ , the coefficient of absorption along the wave is proportional to  $\omega^{6/7}$ .

2. Equation (4.5) is simplified also in the case of large fluctuations but weak spatial dispersion. For simplicity, we set down the results for the nondispersive medium. The dispersion equation obtained in (4.5) again has the form of (4.6) (with  $\rho_0 \tilde{Q} = u_s^2$ ) but the random function  $\rho(\mathbf{r})$  is replaced by  $\eta(\mathbf{r}) = \ln[\rho_0 + \rho(\mathbf{r})]$  and, correspondingly,  $\bar{\rho}^2$  and  $W(\mathbf{r} - \mathbf{r}')$  by  $\eta^2$  and  $W^\eta(\mathbf{r} - \mathbf{r}')$ .

3. In the absence of spatial dispersion ( $\rho_0 \tilde{Q}(\mathbf{k}) = u_s^2$ ) the effect on the absorption coefficient and the sound velocity of scattering by the fluctuations can be obtained in general form.<sup>[14,15]</sup> We turn to Eq. (4.6) which, after application of the inverse Fourier transformation to the function  $W^\rho(\mathbf{q} - \mathbf{p})$ , can be written in the form

$$\begin{aligned} q^2 - \left( \frac{\omega}{u_s} \right)^2 - \frac{\bar{\rho}^2}{\rho_0^2} \int \frac{d\mathbf{p}}{(2\pi)^3} \frac{1}{[p^2 - (\omega/u_s)^2]} \\ \times \int d\mathbf{r} e^{i\mathbf{p}\mathbf{r}} \left( q^2 p^2 - q_i q_j \frac{\partial^2}{\partial r_i \partial r_j} \right) \\ \times W^\rho(\mathbf{r}) e^{-i\mathbf{q}\mathbf{r}} = 0. \end{aligned} \quad (4.8)$$

<sup>7)</sup>As is shown in a number of works, <sup>[14,17]</sup> this method of calculation is more accurate than the usual perturbation method, since it essentially renormalizes an interaction described (in the case of electrodynamics) by the dielectric permittivity. According to <sup>[17]</sup>, renormalizations of higher orders can be carried out in a similar fashion.

The absence of spatial dispersion corresponds to thermal fluctuations  $kr_0 \ll 1$ , therefore, after integration over the wave vector, one can expand  $e^{ikr}$  in a series. Owing to the isotropy of the medium, we assume that the correlation function depends only on the modulus of  $r$ . Finally, substituting  $q \rightarrow k$  in the correction term, we have

$$q^2 = \left( \frac{\omega}{u_s} \right)^2 \left[ 1 + \frac{\overline{\rho^2}}{\rho_0^2} \left( k^2 r_0^2 + i \frac{4}{3} k^3 r_0^3 \right) \right] \quad (4.9)$$

where the correlation function  $W^\rho(\mathbf{r})$  is normalized by the condition

$$r_0^2 = \int r W^\rho(\mathbf{r}) d\mathbf{r},$$

where  $\overline{\rho^2} = k_B T / 4\pi b r_0$  for thermal fluctuations.

Thus, in the case  $kr_0 \ll 1$ , the change in the sound velocity brought about by scattering from the fluctuations is proportional to  $\omega^2$ , while the coefficient of absorption along the wave is  $\sim \omega^3$ .

4. Equation (4.9) was obtained for small fluctuations. This result remains valid however for large fluctuations (in the case of small spatial dispersion) but, in correspondence with what was pointed out above, one must replace  $\overline{\rho^2}$  by  $\overline{\eta^2}$ , and the correlation radius  $r_0$  in (4.9) is determined by means of the function  $W^\eta(\mathbf{r})$ .

5. In the case of weak fluctuations, one can forego the condition of stationarity of the fluctuations and assume  $\rho = \rho_0(\mathbf{r}, t)$ , which is important in the critical region, since the appearance of temporal dispersion and corresponding relaxation phenomena are possible.

In this case, it is convenient to apply the method described by the operator relations (4.4)–(4.5) to each of Eqs. (4.3), and the dispersion equation is obtained as a condition of compatibility of the solutions of the two equations for the regular values  $n_0$  and  $\mathbf{v}$ . The dispersion equation here has the form (4.6) but the Fourier components of  $W(\mathbf{r})$  and  $\tilde{Q}(\mathbf{r})$  also depend on the frequency, i.e., in (4.6),  $\tilde{Q} = \tilde{Q}(\mathbf{p} - \mathbf{q}, \omega)$  and  $W = W(\mathbf{p} - \mathbf{q}, \omega)$ . Correspondingly, integration in (4.6) is carried out not only in wave vector space, but also over the frequencies.

## 5. SCATTERING OF ELECTROMAGNETIC WAVES

A. The scattering of electromagnetic waves is described by the correlator of the fluctuations of the dielectric permittivity

$$\overline{\delta\epsilon(\mathbf{r}, t) \delta\epsilon(\mathbf{r}', t')} = \overline{\delta\epsilon^2} W^\epsilon(\mathbf{r} - \mathbf{r}', t - t'),$$

connected with the correlation functions of the hydrodynamic quantities, which are found above with account of spatial dispersion. If we take into

account only scattering from density fluctuations, then

$$\overline{\delta\epsilon^2} W^\epsilon = (\partial\epsilon/\partial\rho)_T^2 \overline{\delta\rho^2} W^\rho.$$

The extinction coefficient per unit solid angle  $dh/d\omega'$  is expressed in terms of  $Q(\mathbf{q}, 0)$  and (3.16), and depends essentially on the spatial dispersion

$$\frac{dh}{d\omega'} = \frac{\omega^4}{(4\pi)^2 c^4} \left( \frac{\partial\epsilon}{\partial\rho} \right)_T^2 \frac{k_B T}{Q(\mathbf{q}, 0)} \sin^2 \theta, \quad (5.1)$$

where  $\mathbf{q} = \mathbf{k} - \mathbf{k}'$  is the change in the wave vector in the scattering process, while, for small change in frequency,

$$q = 2 \frac{\omega}{c} \sin \frac{\theta}{2}$$

( $\theta$  is the angle between  $\mathbf{k}$  and  $\mathbf{k}'$ ). The angle factor in (5.1) corresponds to the case of linearly polarized waves.

We note that for  $Q(\mathbf{q}) = Bq^{3/2}$  the intensity of the scattered light in the immediate vicinity of the critical point is proportional to  $q^{5/2}$ , and not to  $q^2$ . Furthermore, for  $Q(\mathbf{q}) = Bq^{3/2}$ , integration of (5.1) over the angles leads to a finite expression for the total extinction coefficient, while for  $Q(\mathbf{q}) = a + bq^2$ , a logarithmic divergence is obtained at the critical point ( $a = 0$ ), as is well known.<sup>[8]</sup>

In the presence of spatial dispersion, the well known ratio of Landau-Placzek<sup>[8]</sup> is also altered; this ratio,  $I_{\text{doubl}}/I_{\text{tot}} = c_v/c_p$ , describes the ratio of the intensity of the Mandel'shtam-Brillouin doublet  $I_{\text{doubl}}$  to the total intensity of the line  $I_{\text{tot}} = I_{\text{doubl}} + I_{\text{unshift}}$ :

$$\frac{I_{\text{doubl}}}{I_{\text{unshift}}} = \frac{\rho_0 Q(\mathbf{q})}{\rho_0 Q(\mathbf{q}) + \frac{T_0}{c_v \rho_0^2} \left( \frac{\partial\rho}{\partial T} \right)_\rho^2} = \frac{u_T^2(\mathbf{q})}{u_s^2(\mathbf{q})}. \quad (5.2)$$

For weak dispersion, (5.2) transforms to the corresponding formula of Vladimirskii.<sup>[13]</sup>

It follows from (5.2) that, the intensity of the unshifted line is decreased ( $I_{\text{doubl}}/I_{\text{tot}} \rightarrow 1$ ) in the immediate vicinity of the critical point, as  $c_v \rightarrow \infty$ , in the presence of spatial dispersion. In the absence of spatial dispersion, we would have obtained a decrease in the intensity of the doublet ( $c_v/c_p \rightarrow 0$ ).

A more detailed formula for the Rayleigh scattering line shape is obtained from the expression

$$\frac{dh}{d\omega' d\omega'} = \frac{\omega^4}{(4\pi)^2 c^4} \left( \frac{\partial\epsilon}{\partial\rho} \right)_T^2 \overline{\delta\rho^2} W^\rho(\mathbf{q}, \omega - \omega') \sin^2 \theta. \quad (5.3)$$

Thus the investigation of the line shape of the scattered light, determined by (5.1)–(5.3) for different frequencies and angles of observation, is a convenient method for the experimental determination of the values of  $Q(\mathbf{k}, 0)$  and  $Q(\mathbf{k}, \omega)$ .



B. The scattering of electromagnetic waves also leads to an additional damping and dispersion of the mean field associated with the transfer of energy to the fluctuating part of the field, which can distort the scattering picture. The damping of the average field could be described (as was shown by Lifshitz, Kaganov and Tsukernik<sup>[14]</sup>; see also<sup>[15-17]</sup>), with the help of the effective dielectric permittivity  $\epsilon_{lm}^{\text{eff}}$ . Separating the field and the dielectric susceptibility into the regular and fluctuating parts, and substituting them in Maxwell's equation, we get equations of the type (4.4), which are solved by the method given in Sec. 4. The dispersion equation has the form

$$\text{Det} \left| k^2 - (\omega/c)^2 \epsilon_{lm}^{\text{eff}} \right| = 0, \quad (5.4)$$

$$\begin{aligned} \epsilon_{lm}^{\text{eff}} = & \epsilon \delta_{lm} - \overline{\delta \epsilon^2} \left[ \delta_{lm} - \left( \frac{c}{\omega} \right)^2 \frac{k_l k_m}{\epsilon} \right] \int \frac{d\mathbf{q} d\Omega}{(2\pi)^4} \\ & \times \frac{(\omega/c)^2 \delta_{lm} - k_l k_m / \epsilon}{k^2 - (\omega/c)^2 \epsilon} W^\epsilon(\mathbf{k} - \mathbf{q}, \Omega). \end{aligned} \quad (5.5)$$

The dielectric permittivity (5.5), because of the presence of poles in the integrand, is a complex quantity which corresponds to dispersion and absorption of the mean field.

If we write  $\epsilon_{lm}^{\text{eff}} = \epsilon \delta_{lm} + \Delta \epsilon_{lm}^{\text{eff}}$ , then the solution of the dispersion equation (5.4) in the linear approximation in  $\overline{\delta \epsilon^2}$  is

$$k^2 = \frac{\omega^2}{c^2} \left[ \epsilon + \Delta \epsilon_{ii}^{\text{eff}} \left( \omega, \frac{\omega}{c} \sqrt{\epsilon} \right) \right]. \quad (5.6)$$

In the case of stationary fluctuations  $W^\epsilon(\mathbf{k}, \Omega) = 2\pi W^\epsilon(\mathbf{k}) \delta(\Omega)$  and the well known expression for  $\epsilon_{lm}^{\text{eff}}$  follows from (5.5).<sup>[14,15]</sup> This can be established by transforming to the coordinate representation under the integral sign.

C. Let us make some final remarks. As is well known, the growth of density fluctuations near the critical point leads to the result that the fluctuations are correlated over significant distances. This spatial correlation is a consequence of the simple fact that if there is a significant departure of the density at any point from the equilibrium value, the material from neighboring points must enter into this point. The nonlocal coupling arising in this case between the pressure and the density, (2.19) and (3.7), leads to a dependence of the sound velocity on the wave vector  $\mathbf{k}$  (spatial dispersion).

Both the density correlator and the isothermal sound speed are expressed in terms of the kernel  $\tilde{Q}(\mathbf{r}, T - T_c)$  (2.3) of the non-equilibrium part of the free energy and are connected by (2.17) and (3.16), a relation similar to the Feynman relation between the correlation function and the excitation spectrum in liquid helium. Thus the measurement of the

dispersion of the sound speed enables us in principle to determine the excitation spectrum of  $Q(\mathbf{k}, T - T_c)$  near the critical point. Another method of determining the spectrum is to measure the Fourier components of the density correlator (for example, from the wave scattering). The correlators of hydrodynamic quantities with account of spatial dispersion were obtained in Sec. 3.

The determination of the spectrum is of interest in connection with the problem of second-order phase transitions, since the experimentally discovered singularity of the thermodynamic quantities at the transition point means that the spectrum of  $Q(\mathbf{k})$  for  $T = T_c$  must also contain a singularity for  $\mathbf{k} = 0$ , and the expansion  $Q(\mathbf{k}) = a + b k^2$  is invalid. The experimentally observed logarithmic singularities of  $c_v$  at the critical point evidently correspond to the dispersion law  $Q(\mathbf{k}) = a + B k^3/2$  (see footnote 4<sup>1</sup>).

We note that as a consequence of the spatial dispersion, neither the isothermal nor the adiabatic sound speed vanishes at  $T = T_c$  and  $\mathbf{k} \neq 0$ .<sup>8)</sup> The advantage of the acoustic measurements is the possibility of realizing different relations between the sound parameters and the parameters of the material as a function of the degree of approach to the critical point and the selection of the sound frequency which can vary over wide ranges  $10^5 - 10^9$  cps<sup>[18]</sup>.

Valuable information can be obtained in investigations of the angular dependence of small-angle Rayleigh scattering of electromagnetic waves, and also in investigations of the Mandel'shtam-Brillouin doublet, in particular the Landau-Placzek ratio with spatial dispersion (5.2) (see Sec. 5, and also the review of Fabelinskiĭ<sup>[19]</sup>). In this connection, the experiments of Drickamer et al<sup>[20]</sup> are of interest, in which regions were discovered with different dependences of the scattered light intensity on the wave length, depending on the approach to the critical point.

The value of the sound absorption and, consequently, the possibility of acoustic experiments, depends essentially on the behavior close to the

<sup>8)</sup>The few existing experiments on the propagation of sound in the critical region<sup>[18]</sup> show that on approaching the critical point the absorption increases while the sound speed decreases. These experiments were carried out under conditions of weak spatial dispersion, i.e., the decrease of sound speed is approximately determined by the relation (1.4)  $u_s \sim 1/\sqrt{c_v}$ , and since when the distance to the critical point changes by two orders of magnitude (from  $\Delta T = 1^\circ$  to  $\Delta T = (10^{-2})^\circ$ ) the heat capacity of argon increases by a factor of four, the sound speed decreases only by a factor of two.

critical point of the thermal conductivity and the viscosity, which have not been studied to date.

In addition, in a medium with strongly fluctuating parameters, there is a specific mechanism of absorption of waves associated with the transfer of energy to the fluctuating part of the field (Sec. 4B, C and Sec. 5B), which can lead to distortion of the usual scattering picture.

<sup>1</sup>L. D. Landau and E. M. Lifshitz, *Statisticheskaya fizika (Statistical Physics)*, Gostekhizdat, 1951.

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