[11]V. A. Lyul'ka, Zh. Eksp. Teor. Fiz. **67**, 1638 (1974) [Sov. Phys. JETP **40**, 815 (1975)].
[12]V. A. Lyul'ka, Zh. Eksp. Teor. Fiz. **69**, 800 (1975) [Sov. Phys. JETP **42**, 408 (1975)].
[13]V. N. Baĭer, V. M. Katkov, A. I. Mil'shteĭn, and V. M. Strakhovenko, Zh. Eksp. Teor. Fiz. **69**, 783 (1975) [Sov. Phys. JETP **42**, 400 (1975)].
[14]V. B. Berestetskiĭ, E. M. Lifshitz, and L. P. Pitaevskiĭ, Relyativistskaya kvantovaya teoriya, ch. 1 (Relativistic quantum theory, part 1), Fizmatgiz, 1968 (Engl. Transl. Pergamon, N.Y., 1971).
[15]J. A. Giordmaine and R. C. Miller, Phys. Rev. Lett. **14**, 973 (1965).
[16]S. A. Akhmanov, A. I. Kovrigin, A. S. Piskarskas, V. V. Fadeev, and R. V. Khokhlov, Pis'ma Zh. Eksp. Teor. Fiz. **2**, 300 (1965) [JETP Lett. **2**, 191 (1965)].
[17]A. D. White and J. D. Rigden, Proc. IEEE **51**, 943 (1963); H. J. Gerritsen and P. V. Goedertier, Appl. Phys. Lett. **4**, 20 (1964); D. Rosenberger, Phys. Lett. **9**, 29 (1964); R. Der Agobian, Phys. Lett. **10**, 64 (1964).
[18]Amnon Yariv, Quantum electronics, Wiley, N. Y., 1967 and 1975 [Russ. Transl. Kvantovaya elektronika i nelineĭnaya optika (Quantum electronics and nonlinear optics), Sovetskoe radio, 1973].
[19]V. M. Arutyunyan, E. G. Kanetsyan, and V. O. Chaltynyan, Zh. Eksp. Teor. Fiz. **59**, 195 (1970) [Sov. Phys. JETP **32**, 108 (1971)].
[20]M. V. Fedorov, Zh. Eksp. Teor. Fiz. **68**, 1209 (1975) [Sov. Phys. JETP **41**, 601 (1975)].
[21]I. V. Lebedev, Opt. Spektrosk. **29**, 948 (1970) [Opt. Spectrosc. (USSR) **29**, 503 (1970)].
[22]V. Ch. Zhukovskiĭ and I. Kherrmann, Vestnik MGU, seriya fiz., **6**, 671 (1970).
[23]A. I. Nikishov and V. I. Ritus, Zh. Eksp. Teor. Fiz. **52**, 1707 (1967) [Sov. Phys. JETP **25**, 1135 (1967)].
[24]A. I. Nikishov and V. I. Ritus, Zh. Eksp. Teor. Fiz. **46**, 1768 (1968) [Sov. Phys. JETP **19**, 1101 (1964)].
[25]L. S. Brown and T. W. B. Kibble, Phys. Rev. **133**, A705 (1964).

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Multiple scattering of light in an inhomogeneous medium near the critical point. II. Spectral composition of scattered radiation

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The spectral composition of light after multiple scattering in an inhomogeneous medium is examined. The number of components, the positions of their centering points, the half widths, the relationships between the components, and the characteristics of the components in the spectra are determined. Conditions for mode conservation and decay during multiple scattering are formulated. The "field" and temprature dependence of the properties of the single- and double-scattering spectra near the critical point are described. The dependence of these spectra on the transferred wave vector is also discussed. An experiment designed to investigate multiparticle correlations in a condensed medium is outlined.

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Multiple scattering of light near the critical point has been attracting increasing attention among researchers. The theory of integrated multiple scattering of light in the critical region has now been developed in considerable detail.[1–3] Thus, among the new and interesting results of these studies is the prediction[1,2] and discovery[4,5] of a nontrivial dependence of the doubly-scattered intensity on the fourth power of the linear size of the scattering volume; the gradual reduction of the exponent in the temperature dependence of doubly scattered radiation[1,6] as the critical point is approached is in agreement with theoretical predictions based on the theory of scaling transformations[7]; the role of external factors in the description of critical opalescence has been elucidated[8–11]; the properties of the scattering indicatrix in the critical region has been investigated[1,2,9,11]; and so on. Considerable success has also been achieved in the study of the depolarization of critical opalescence.[12,13,4–6]

Higher-order scattering effects are just as important for the spectra of critical opalescence. Unfortunately, a theory of multiply scattered spectra, capable of predicting and explaining their experimentally accessible properties, is not as yet available. In this paper, we solve this problem for the liquid-vapor critical point. In addition to the study of temperature dependence, which is now traditional in the investigation of critical opalescence, we have here the further possibility of examining the "field" properties connected with departures from the homogeneity of the medium under the influence of external factors (for example, the gravitational field). This was used in our earlier work on the physical properties of such inhomogeneous media near the critical point in the single-scattering approximation.[8,9,14]

We shall devote particular attention to the number of components in spectra of different order, the position of the centering points, the half-widths, and the relationships between the components in the spectra, as well as the temperature, "field," and angular dependence of the multiply-scattered spectra. The general

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results obtained below will be useful for the description not only of the critical opalescence spectra, but also in the study of the spectral composition of multiply-scattered radiation in turbid media of arbitrary nature.

1. In an optically inhomogeneous turbid medium with $\mu=1$, $\varepsilon(\mathbf{r},t)=\varepsilon_0(\mathbf{r})+\varepsilon'(\mathbf{r},t)$, the scattering of electromagnetic waves is described by the following set of coupled integral equations:

$$\mathbf{E}_i(\mathbf{r},t)=\int_V\int_0^\infty G^{(-)}(\mathbf{r},\mathbf{r}_i,t-t_i)\{\Phi_{i-1}(\mathbf{r}_i,t_i)-\nabla_i(\mathbf{E}_i(\mathbf{r}_i,t_i)\nabla_i\ln\varepsilon_0(\mathbf{r}_i))\}d\mathbf{r}_i\,dt_i, \tag{1}$$

$$\mathbf{H}_i(\mathbf{r},t)=-c\int_0^\infty\theta(t-t_k)[\nabla\mathbf{E}_i(\mathbf{r},t_k)]dt_k \tag{2}$$

with initial conditions

$$\mathbf{E}_i(\mathbf{r},t)|_{t\leqslant t_i}=0,\quad \frac{\partial\mathbf{E}_i(\mathbf{r},t)}{\partial t}\Big|_{t\leqslant t_i}=0,\quad i=1,2,3,\ldots \tag{3}$$

In these expressions,

$$\Phi_{i-1}=\frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\varepsilon'\mathbf{E}_{i-1})-\nabla\left(\mathbf{E}_{i-1}\frac{\nabla\varepsilon'}{\varepsilon_0}\right),$$

$\varepsilon_0(\mathbf{r})$ and $\varepsilon'(\mathbf{r},t)$ are, respectively, the macroscopic and fluctuation components of scalar permittivity, and $G^{(-)}(\mathbf{r},\mathbf{r}_i,t-t_i)$ is the causal Green function of the differential operator in the wave equation that is equivalent to (1). In the approximation of continuous inhomogeneity,[9] the latter operator is given by

$$G_0^{(-)}(\mathbf{r},\mathbf{r}_i,t-t_i)=-\frac{\theta(t-t_i)}{4\pi|\mathbf{r}-\mathbf{r}_i|}\delta\left\{(t-t_i)-\frac{\varepsilon_0^{1/2}(\mathbf{r}_i)|\mathbf{r}-\mathbf{r}_i|}{c}\right\}, \tag{4}$$

where

$$\theta(t-t_i)=\begin{cases}1, & t>t_i\\ 0, & t<t_i\end{cases}$$

is a step function that orders the scattering events in time.

If we solve (1)–(3) with the Green function given by (4), we obtain the following recurrence relations for the fields after i-th scatter at $\mathbf{r}$ in the ω representation:

$$\mathbf{E}_i(\mathbf{r},\omega_i)=-\pi\int_V\int_{-\infty}^\infty\varepsilon'(\mathbf{r}_i,\omega)G_0^{(-)}(\mathbf{r},\mathbf{r}_i,\omega_i)\left\{\frac{\omega_i^2}{c^2}\mathbf{E}_{i-1}(\mathbf{r}_i,\omega-\omega_i)\right.$$
$$\left.+\left[1-i\frac{\omega_i}{c}\varepsilon_0^{1/2}(\mathbf{r}_i)|\mathbf{r}-\mathbf{r}_i|-\frac{\omega_i^2}{c^2}\varepsilon_0(\mathbf{r}_i)|\mathbf{r}-\mathbf{r}_i|^2\right.\right.$$
$$\left.\left.\times\frac{\mathbf{n}_i(\mathbf{n}_i\mathbf{E}_{i-1}(\mathbf{r}_i,\omega-\omega_i))}{\varepsilon_0(\mathbf{r}_i)|\mathbf{r}-\mathbf{r}_i|^2}\right]\right\}d\mathbf{r}_i\,d\omega, \tag{5}$$

$$\mathbf{H}_i(\mathbf{r},\omega_i)=\frac{i\pi\omega_i}{c}\int_V\int_{-\infty}^\infty\varepsilon'(\mathbf{r}_i,\omega)G_0^{(-)}(\mathbf{r},\mathbf{r}_i,\omega_i)$$
$$\times\left[1+i\frac{\omega_i}{c}\varepsilon_0^{1/2}(\mathbf{r}_i)|\mathbf{r}-\mathbf{r}_i|\right][\mathbf{n}_i\mathbf{E}_{i-1}(\mathbf{r}_i,\omega-\omega_i)]d\mathbf{r}_i\,d\omega, \tag{6}$$

where ω_i is the frequency of i-tiply scattered light.

We have used the WKB criterion [see Eq. (1.3)][1)] in the solution of (1)–(3). For the geometry used in[1,9], this enables us to write the solution corresponding to an exciting wave $\mathbf{E}_0$ at frequency ω_0 in the form

$$\mathbf{E}_0(z,\omega)=\mathbf{m}_0E_0(z)\delta(\omega-\omega_0). \tag{7}$$

Analysis of multiple scattering near the critical point cannot be carried out unless the space-time correlation functions $g_{l+m}(\mathbf{r}_1,t_1,\ldots,\mathbf{r}_l,t_l;\mathbf{r}_l',t_1',\ldots,\mathbf{r}_m',t_m')$, $l+m>2$ are known for the density fluctuations. "Pure" scattering effects are described by space-time correlation functions of the same order, whereas "interference" effects are described by correlation functions of different order [see (1.10)]. The explicit form of this space-time correlation function for $l+m>2$ in the neighborhood of the critical point is not as yet known. However, the wave zone condition (1.7), which selects periodic solutions for $\mathbf{E}_i$ and $\mathbf{H}_i$ after the successive scattering events, can be used to determine the main contributions to the spectrum of pure i-tiple scattering with the aid of the pair space-time correlation function for an inhomogeneous medium, found previously in[15] with allowance for memory effects and the spatial dispersion of kinetic coefficients in the nonlocal hydrodynamic equations, which is necessary near the critical point.

The contribution of interference effects to the critical opalescence spectrum is, unfortunately, difficult to determine because the precise asymptotic form of the space-time correlation functions of odd order, which corresponds to the wave zone condition, is not known. Nevertheless, it is possible to obtain the shape of the spectrum and its most important quantitative characteristics (with the exception of the integrated intensity of the individual components) by using a dynamic analog of the static calculation of the interference terms in the Poynting-Umov vector $\langle\mathbf{S}_{ii}\rangle$.[1]

One of the consequences of the analysis performed in[15] is that the general expression for the pair space-time correlation function $g(\mathbf{k},\mathbf{k}',\omega)$ in a spatially inhomogeneous medium, in which temporal nonlocality of the coefficients of the hydrodynamic equations can be neglected, gives the simple three-pole system

$$\omega^{(1,2)}(k)=\pm\operatorname{Re}f(k)-i\operatorname{Im}f(k),\quad \omega^{(3)}=-i\operatorname{Im}\varphi(k), \tag{8}$$

and, if we retain the main contributions in (8),

$$\operatorname{Re}f(k)=k\left(\frac{\partial p}{\partial T}\right)_\rho^{1/2}\left\{\left[\left(\frac{\partial e}{\partial\rho}\right)_T+h\right]\left[\frac{1}{c_V(z,\tau)}+B_ck^2\right]\right\}^{1/2}, \tag{9}$$

$$\operatorname{Im}f(k)=\frac{1}{2}k^2\left\{\left(\frac{1}{\zeta(z,t)}+B_\zeta k^2\right)^{-1}+\left(\frac{3}{4\eta(z,\tau)}+B_\eta k^2\right)^{-1}-\operatorname{Im}\varphi(k)\right\}, \tag{10}$$

$$\operatorname{Im}\varphi(k)=k^2\left[\left(\frac{\partial p}{\partial\rho}\right)_T+c_\rho^{(2)}k^2\right]\left[\left(\frac{\partial p}{\partial T}\right)_\rho\right.$$
$$\left.\times\left(\left(\frac{\partial e}{\partial\rho}\right)_T+h\right)\left(\frac{1}{\lambda(z,\tau)}+B_\lambda k^2\right)\right]^{-1}. \tag{11}$$

These expressions are written in standard notation.[15,16] The terms B_ik^2 are added to the quantities which vanish at the critical point in order to take into account spatial dispersion of the fluctuations ($c_\rho^{(2)}\to B_\rho$).

To estimate the singularities in (9)–(11), we must use the formulas of the theory of similarity[7,15]

$$\varphi_i(z,\tau)=\varphi_i^{(0)}(T_c,\rho)+m_i|z|^{-\varepsilon_i/\beta\delta}\left(1+n_i\frac{\tau}{|z|^{1/\beta\delta}}+\ldots\right) \tag{12}$$

$$\varphi_i(z,\tau)=\varphi_i^{(0)}(T,\rho_c)+s_i|\tau|^{-\varepsilon_i}\left(1+l_i\frac{z^2}{\tau^{2\beta\delta}}+\ldots\right) \tag{13}$$

which are valid in the neighborhood of the critical isotherm and critical isochore, respectively. The exponents ε_i assume the values $\alpha \approx 0.1-0.2$ for the specific heat at constant volume c_V and, apparently, shear viscosity η; $3\nu+\alpha=2$ for second viscosity ζ; $\gamma-\nu \approx 2/3$ for the thermal conductivity λ; $\gamma \approx 1.2-1.3$ for the derivative $(\partial p/\partial \rho)_t$; $\varphi_i^{(0)}$ are the regular parts of the corresponding kinetic coefficients; $\tau=(T-T_c)/T_c$; m_i, n_i, s_i, and l_i are constant coefficients.

To facilitate our calculations and to obtain manageable expressions for the spectral intensities of multiply scattered light, it will be convenient to introduce the following notation.

1) The contribution to the spectrum connected with the thermal mode root $\omega^{(3)}$ is determined by the quantity

$$M^0=\left(1-\frac{2\Delta'}{\Delta}\right)\mathcal{L}(\Omega_i, 0, \operatorname{Im}\varphi);$$

where

$$\Delta=2\operatorname{Re} f[(\operatorname{Re} f)^2+(\operatorname{Im} f-\operatorname{Im}\varphi)^2],$$
$$\Delta'=\operatorname{Re} f[\Delta_1+(\operatorname{Im} f-\operatorname{Im}\varphi)\Delta_2],$$
$$\Delta_1=(\operatorname{Re} f)^2-(\operatorname{Im} f)^2+a_1\operatorname{Im} f-a_2,\quad \Delta_2=2\operatorname{Im} f-a_1,$$
$$\mathcal{L}(\Omega, \pm u, v)=\frac{1}{\pi}\frac{v}{(\Omega\mp u)^2+v^2}$$

is the normalized Lorentzian whose parameters u and v determine the real and imaginary parts of the roots in (9)–(11).

2) The contributions to the spectrum connected with the acoustic-mode roots $\omega^{(1)}$ and $\omega^{(2)}$ are determined by

$$M^+=\frac{\Delta'}{\Delta}\mathcal{L}(\Omega_i, \operatorname{Re} f, \operatorname{Im} f),\qquad M^-=\frac{\Delta'}{\Delta}\mathcal{L}(\Omega_i, -\operatorname{Re} f, \operatorname{Im} f).$$

3) The number of subscripts (0, +, −) corresponds to the scattering multiplicity in the particular approximation. It determines the total contribution of the real and imaginary parts of the roots (8) to u and v and, at the same time, represents the dependence of the Lorentzian on the transferred frequency $\Omega_i=\omega_i-\omega_0$.

4) The index number, counted from the left, indicates its dependence on the corresponding transferred wave vector in successive scattering events.

Let us illustrate these definitions by the following example;

$$M^{0-+}=\left(1-\frac{2\Delta'(\mathbf{q}_1)}{\Delta(\mathbf{q}_1)}\right)\frac{\Delta'(\mathbf{q}_2)\Delta'(\mathbf{q}_3)}{\Delta(\overline{\mathbf{q}}_2)\Delta(\mathbf{q}_3)}\mathcal{L}[\Omega_3, -\operatorname{Re} f(\mathbf{q}_2)+\operatorname{Re} f(\mathbf{q}_3),$$
$$\operatorname{Im}\varphi(\mathbf{q}_1)+\operatorname{Im}(f(\mathbf{q}_2)+f(\mathbf{q}_3))].$$

The centering point $\Omega_{c,m}$ of the m-th component of the i-tiply scattered spectrum along the frequency axis will be determined from the following equation:

$$\operatorname{Im}\int_0^\infty x\left[\int_{\sigma_1}\cdots\int_{\sigma_{i-1}}\exp[-iu(\sigma_1,\ldots,\sigma_{i-1})x-v(\sigma_1,\ldots,\sigma_{i-1})x]\prod_{k=1}^{i-1}d\sigma_k\right]$$
$$\times\exp(i\Omega_{c,m}x)\,dx=0. \tag{14}$$

The half-widths Γ_m of the components in the spectra will be defined as follows:

$$\frac{1}{2}\int_{\sigma_1}\cdots\int_{\sigma_{i-1}}\frac{1}{v(\sigma_1,\ldots,\sigma_{i-1})}\prod_{k=1}^{i-1}d\sigma_k$$
$$=\int_{\sigma_1}\cdots\int_{\sigma_{i-1}}\frac{v(\sigma_1,\ldots,\sigma_{i-1})}{\Gamma_m^2+v^2(\sigma_1,\ldots,\sigma_{i-1})}\prod_{k=1}^{i-1}d\sigma_k. \tag{15}$$

This expression is the formal generalization of the commonly accepted definition of the half-width Γ at half-height in singly-scattered spectra, namely,

$$^1/_2 J_{max}=J(\Gamma).$$

2. When the critical opalescence spectrum is investigated in the single-scattering approximation, the differential of the pure-scattering spectral intensity with respect to height can be obtained with the aid of (5)–(7) with $i=1$:

$$\frac{dI_1(\Omega_1)}{dz}=\frac{dI_1^{\text{stat}}}{dz}\{M^0+M^++M^-\}, \tag{16}$$

where

$$\frac{dI_1^{\text{stat}}}{dz}=\int_{-\infty}^{\infty}\frac{dI_1(\Omega_1)}{dz}d\Omega_1$$

is the static part (integrated intensity) which is given by (1.15). The first term in (16) is the unshifted component (thermal mode) and the other two form the Mandel'shtam-Brillouin doublet (acoustic modes).

It is readily shown that the following natural conservation condition is satisfied:

$$\int_{-\infty}^{\infty}\{M^0+M^++M^-\}d\Omega_1=1. \tag{17}$$

Hence, it follows directly that the ratio of the integrated intensities of the fine-structure component in the singly scattered spectrum is

$$\frac{J_c}{2J_{MB}}=\frac{\Delta(q_1)}{2\Delta'(q_1)}-1,\qquad \mathbf{q}_1=\frac{\varepsilon_0^{1/2}}{c}\omega_0(\mathbf{n}_1-\mathbf{n}_0). \tag{18}$$

This expression becomes identical with the well-known Landau-Placek relation when the space-time dispersion is neglected well away from the critical points.

The centering points and half-widths of the components of the singly-scattered spectrum are given in our notation by

$$\Omega_c=0,\quad \Omega_{MB}=\pm\operatorname{Re} f(q_1;\tau,z), \tag{19a}$$
$$\Gamma_c=\operatorname{Im}\varphi(q_1;\tau,z),\quad \Gamma_{MB}=\operatorname{Im} f(q_1;\tau,z), \tag{19b}$$

which yields the well-known results for the homogeneous system near the critical point.[16,17] In the usual experimental situation, where the effect of the external field cannot be neglected, the characteristics (19) of the singly-scattered triplet are given by (9)–(13). In particular, near the critical isotherm ($|\tau|^{\beta\delta}\ll z$), the singly-scattered spectrum depends on height, i.e., for a fixed angle of observation, the centering points of the Mandel'shtam-Brillouin components depart from the central line ($\Omega_{MB}\sim|z|^{\alpha/\beta\delta}$) as $|z|$ increases, i.e., with

increasing distance from the level with maximum density gradient and, at the same time, the half-width of the component decreases ($\Gamma_{MB} \sim |z|^{-(3\nu+\alpha)/\beta\delta}$) whereas the half-width of the central component increases ($\Gamma_c \sim |z|^{(\gamma-\nu)/\beta\delta}$).[15]

The contribution of the first-order interference terms to the singly-scattered spectrum is zero because the linear fluctuation in permittivity is zero.

3. Calculation of the spectral intensity of pure double scattering differentiated with respect to height and based on (5)–(6) with $i=2$, the wave zone condition (1.7), and the space-time correlation function $g(\mathbf{k}, \mathbf{k}', \omega)$ obtained in[15], which is necessary for the determination of the main contribution to the four-point space-time correlation function, yields the following result after integration with respect to the transferred frequency $\Omega_1(\Omega_1/\omega_0 \ll 1)$ and some quite complicated transformations:

$$\frac{dI_2(\Omega_2)}{dz} = \int_{\sigma_1} \frac{d^2 I_2^{\text{стат}}}{dz\, d\sigma_1} \{M^{00} + M^{0+} + M^{0-} + M^{+0} + M^{++} + M^{+-} + M^{-0} + M^{-+} + M^{--}\} d\sigma_1, \tag{20}$$

where

$$\frac{dI_2^{\text{стат}}}{dz} = \int_{-\infty}^{\infty} \frac{dI_2(\Omega_2)}{dz} d\Omega_2$$

is the integrated pure doubly-scattered intensity given by (1.19), and the integral with respect to solid angle gives the sum along the given direction of all the singly-scattered intensities in the volume under investigation, which are responsible for the doubly-scattered radiation intercepted by the detector.

The various terms in (20) correspond to the different physical processes occurring as a result of two successive scattering events. The first three describe the redistribution of the central thermal mode of single scattering into the unshifted thermal and two frequency-shifted acoustic modes of double scattering. The last two triplets of terms correspond to the redistribution of the right and left components of Mandel'shtam-Brillouin single scattering within the volume into unshifted thermal and shifted acoustic modes of double scattering. Using the rules given by 3) and 4), it can be shown that these terms are given by

$$M^{00} = \left[1 - \frac{2\Delta'(q_1)}{\Delta(q_1)}\right]\left[1 - \frac{2\Delta'(q_2)}{\Delta(q_2)}\right] \mathcal{L}[\Omega_2, 0, \operatorname{Im}(\varphi(q_1) + \varphi(q_2))], \tag{21}$$

$$M^{0+}, M^{0-} = \left[1 - \frac{2\Delta'(q_1)}{\Delta(q_1)}\right] \frac{\Delta'(q_2)}{\Delta(q_2)} \mathcal{L}[\Omega_2, \pm \operatorname{Re} f(q_2), \operatorname{Im}(\varphi(q_1) + f(q_2))], \tag{22}$$

$$M^{+0}, M^{-0} = \frac{\Delta'(q_1)}{\Delta(q_1)} \left[1 - \frac{2\Delta'(q_1)}{\Delta(q_1)}\right] \mathcal{L}[\Omega_2, \pm \operatorname{Re} f(q_1), \operatorname{Im}(f(q_1) + \varphi(q_2))], \tag{23}$$

$$M^{++}, M^{--} = \frac{\Delta'(q_1)\Delta'(q_2)}{\Delta(q_1)\Delta(q_2)} \mathcal{L}[\Omega_2, \pm \operatorname{Re}(f(q_1) + f(q_2)), \operatorname{Im}(f(q_1) + f(q_2))], \tag{24}$$

$$M^{+-}, M^{-+} = \frac{\Delta'(q_1)\Delta'(q_2)}{\Delta(q_1)\Delta(q_2)} \mathcal{L}[\Omega_2, \pm \operatorname{Re}(f(q_1) - f(q_2)), \operatorname{Im}(f(q_1) + f(q_2))]. \tag{25}$$

The decay of each of the single-scattering modes, which occurs during the second scattering event, is subject to the following conditions:

$$\int_{-\infty}^{\infty} \{M^{00} + M^{0+} + M^{0-}\} d\Omega_2 = 1 - \frac{2\Delta'(q_1)}{\Delta(q_1)}, \tag{26}$$

$$\int_{-\infty}^{\infty} \{M^{+0} + M^{++} + M^{+-}\} d\Omega_2 = \int_{-\infty}^{\infty} \{M^{-0} + M^{-+} + M^{--}\} d\Omega_2 = \frac{\Delta'(q_1)}{\Delta(q_1)}. \tag{27}$$

These conditions yield the conservation condition for double scattering, which is the analog of (17);

$$\int_{-\infty}^{\infty} \left\{\begin{matrix}\text{sum of all} \\ \text{double modes}\end{matrix}\right\} d\Omega_2 = 1. \tag{28}$$

The centering points and half-widths in (21)–(25) in the case of "pure" doubly-scattered intensity (20) in the limit as $k \to 0$ can be obtained from (14) and (15):

Modes:	M^{00}	M^{0+}	M^{0-}	M^{+0}	M^{++}	M^{+-}	M^{-0}	M^{-+}	M^{--}
Centering points Ω_c:	0	$\Omega_c^{(1)}$	$-\Omega_c^{(1)}$	$\Omega_c^{(1)}$	$\Omega_c^{(2)}$	0	$-\Omega_c^{(1)}$	0	$-\Omega_c^{(2)}$
Half-widths Γ:	$\Gamma^{(1)}$	$\Gamma^{(2)}$	$\Gamma^{(2)}$	$\Gamma^{(2)}$	$\Gamma^{(3)}$	$\Gamma^{(3)}$	$\Gamma^{(2)}$	$\Gamma^{(3)}$	$\Gamma^{(3)}$

In the foregoing discussion, we have used the following notation:

$$\Omega_c^{(1)} = h_2 (2\varepsilon_0)^{1/2} \frac{\omega_0}{c}, \quad \Omega_c^{(2)} = 2^{1/2} h_2 (2\varepsilon_0)^{1/2} \frac{\omega_0}{c} (1 + \cos \widehat{\mathbf{n}_2 \mathbf{n}_0})^{1/2}, \quad 0 \leqslant \widehat{\mathbf{n}_2 \mathbf{n}_0} \leqslant \frac{\pi}{2}, \tag{29}$$

$$\Gamma^{(1)} = 2h_1 \varepsilon_0^{1/2} \frac{\omega_0}{c} q_2^n, \quad \Gamma^{(2)} = 2(h_1 h_3 \varepsilon_0)^{1/2} \frac{\omega_0}{c} q_2^n, \quad \Gamma^{(3)} = 2h_3 \varepsilon_0^{1/2} \frac{\omega_0}{c} q_2^n, \tag{30}$$

where

$$h_1 = \frac{\operatorname{Im} \varphi}{k^2}, \quad h_2 = \frac{\operatorname{Re} f}{k}, \quad h_3 = \frac{\operatorname{Im} f}{k^2} \tag{31}$$

are coefficients which are independent of k and $q_2^n = (2\varepsilon_0)^{1/2} \omega_0 c^{-1} (1 - \cos \widehat{\mathbf{n}_2 \mathbf{n}_0})^{1/2}$ is the total wave vector transferred during double scattering through the angle $\vartheta = \widehat{\mathbf{n}_2 \mathbf{n}_0}$.

In accordance with the foregoing results, the experimentally observed fine structure of the pure doubly-scattered spectrum should have the form of the quintet, shown schematically in Fig. 1.

The central component L_0 is triply degenerate. It is made up of the following three terms:

$$(L_0) = M^{00} + M^{+-} + M^{-+}. \tag{32}$$

This is the origin of the line shape of this component. The doubly degenerate side components $+L_1$ and $-L_1$ consist of the following terms:

$$(+L_1) = M^{0+} + M^{+0}, \quad (-L_1) = M^{0-} + M^{-0}. \tag{33}$$

The extreme components

$$(+L_2) = M^{++}, \quad (-L_1) = M^{--} \tag{34}$$

are not degenerate.

The structure of the spectrum of pure doubly-scattered radiation described above can also be obtained

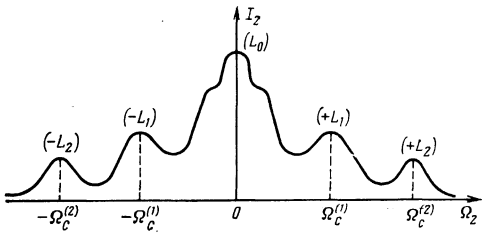

FIG. 1.

from simple qualitative considerations and without using the above data on the centering points Ω_c of the individual components. For M^{0+} and M^{+0}, the symmetry of the corresponding integrals with respect to the solid angle in (14) with $i=2$ under the interchange $\mathbf{n}_2 \rightleftarrows \mathbf{n}_0$ shows that their positions along the frequency axis are identical. For the same reason, M^{+-} and M^{-+} are centered on the same frequency. It is readily seen that M^{++} is centered on a frequency not equal to the centering frequency of either M^{+0} or M^{+-}. Since the spectrum should be symmetric with respect to the position of the double thermal mode M^{00}, the foregoing argument leads immediately to a quintet in the spectrum of pure doubly-scattered radiation.

It is clear that the half-width of the side components $(\pm L_1)$ is $\Gamma^{(2)}$ and that of the extreme components $(\pm L_2)$ is $\Gamma^{(3)}$. It follows from (32) that the half-width of the central component (L_0) is determined by the superposition of the half-widths of the double thermal and two acoustic modes, i.e., $\Gamma^{(1)}$ and $\Gamma^{(3)}$, so that the interpretation of the pattern is quite complicated.

We emphasize the interesting point that, in contrast to singly scattered radiation, for which the half-widths of the fine-structure components of the Rayleigh line are proportional to the square of the transferred wave vector q^2 in the hydrodynamic limit [see (19), (10), and (11)], in the approximation of pure double scattering, the half-widths of all the spectrum components are proportional to the *first* power of q [see (30)]. Moreover, as in the case of the single-scattering approximation, the half-widths of the spectrum components in the case of double scattering tend to zero, but more slowly, in the limit of zero angle scattering $(\vartheta \to 0)$. Thus, the ratio of the half-width Γ_{MB} of the Mandel'shtam-Brillouin component of single scattering to the half-width $\Gamma^{(3)}$ of the extreme components $(\pm L_2)$ of the doubly scattered radiation is given by

$$\Gamma_{MB}/\Gamma^{(3)}=2^{-1/2}(1-\cos\vartheta)^{1/2}, \text{ i.e., } 0\leqslant\Gamma_{MB}/\Gamma^{(3)}\leqslant 1. \tag{35}$$

This formula reflects the fact that an additional broadening of the spectrum components of multiply scattered radiation occurs during mode decay in the course of successive scattering events.

An important consequence of (30) is the following exact relationship between the half-widths $\Gamma^{(i)}$ $(i=1, 2, 3)$:

$$(\Gamma^{(2)})^2=\Gamma^{(1)}\Gamma^{(3)}. \tag{36}$$

Analysis of the function $\Omega_c^{(i)}(q_2^n)$ given by (29) will show that the components $\pm L_1$ remain fixed as the angle of observation ϑ is varied, but the components $\pm L_2$ exhibit a weaker [as compared with the single-scattering approximation; see (9) and (19)] dependence on the transferred wave vector, namely,

$$\Omega_{c(\pm L_2)}=\pm\Omega_c^{(2)}(q_2^n)=\pm 2\Omega_c^{(1)}\left\{1-\frac{1}{2}\left[2\varepsilon_0\left(\frac{\omega_0}{c}\right)^2\right]^{-1}(q_2^n)^2\right\}^{1/4}. \tag{37}$$

In contrast to single scattering, for which the spectrum shrinks to $\Omega=0$, the spectrum of pure doubly scattered radiation at zero scattering angle is still in the form of a quintet and, moreover, $\Omega_c^{(2)}(\vartheta=0)=2\Omega_c^{(1)}$. This fact is connected with the presence of the integral with respect to the solid angle σ_1 which, of course, is absent in the case of single scattering. A situation analogous to that prevailing in single scattering through zero angle is observed in doubly-scattered radiation only for $\tilde{\mathbf{n}}_1=\mathbf{n}_0=\mathbf{n}_2$. However, it is precisely the direction $\tilde{\mathbf{n}}_1$ that makes the zero contributions to the integrals in (14) for all terms with $i=1$. All other directions $\tilde{\mathbf{n}}_1$ give a nonzero result for the terms in the components $\pm L_1$ and $\pm L_2$, and this ensures that the spectrum components are not reduced for $\vartheta \to 0$.

The main contribution to second-order interference effects are provided by the terms in (1.23) containing the three-particle space-time correlation functions because the correlator $\langle[\mathbf{E}_0\times\mathbf{H}_2^*]\rangle+\langle[\mathbf{E}_2\times\mathbf{H}_0^*]\rangle$ [see (1.10) for $i=2$] turns out to be negligible. [1]

Examination of the three-particle space-time correlation function in the wave-zone approximation (1.7) on the basis of the dynamic analog of the well-known asymptotic formulas given in [18-20], in which the factor in front of the pair space-time correlation function can be looked upon as a functional of the mean density, leads to the following expression for the spectral intensity of second-order interference effects differentiated with respect to height:

$$\frac{dI_{1/2}(\Omega_1)}{dz}=\int_{\sigma_1}\frac{d^2I_{1/2}^{\text{stat}}}{dz\,d\sigma_1}\{M^0+M^++M^-\}d\sigma_1, \tag{38}$$

where $dI_{3/2}^{\text{stat}}/dz$ is defined in (1.24). Analysis of (38) will show that the fine-structure components in the spectrum of second-order interference effects take the form of three δ-shaped peaks ("frozen" triplet) with half-widths $\Gamma_c^{(3/2)}=0$ and $\Gamma_{MB}^{(3/2)}=0$, centered on the frequencies $\Omega_c^{(3/2)}=0$, $\Omega_{MB}^{(3/2)}=\pm 2\Omega_c^{(1)}$. As in the static case, the contribution of $dI_{3/2}(\Omega_1)/dz$ to the spectral intensity is substantially smaller than the contribution of $dI_2(\Omega_2)/dz$.

It is clear from (9), (12), (13), (29), and (31) that the field and temperature dependence of the centering points in the doubly scattered spectrum at a given angle of observation are precisely the same as in the single-scattering approximation, i.e.,

$$\Omega_c^{(1)}, \Omega_c^{(2)} \sim h_2 = |\tau|^{\alpha/2}\Psi(z/\tau^{\beta\delta}),$$

where the scaling function $\Psi(x)$ has the following well-known asymptotic behavior:

$$\Psi(x\to 0)=\text{const}, \quad \Psi(x\to\infty)\sim x^{\alpha/2\beta\delta}.$$

The half-widths $\Gamma^{(1)}$ and $\Gamma^{(2)}$ of the double thermal and double acoustic modes are shown by (10)–(13), (30), and (31) to have the same properties as the half-widths Γ_c and Γ_{MB}, given by (19), respectively. The half-width $\Gamma^{(2)}$ for the mixed modes can readily be found from (36):

$$\Gamma^{(2)} \sim |\tau|^{(\gamma-4\nu+\alpha)/2} \Psi(z/\tau^{\beta\delta}).$$

All the above half-widths, i.e., Γ_c, Γ_{MB}, and $\Gamma^{(i)}$ $(i = 1, 2, 3)$ are completely determined by the singular behavior of h_1 and h_3, which are connected with the kinetic coefficients of temperature diffusivity and the second and shear viscosities in the critical region. Moreover, it can be shown theoretically that the latter exhibits a weak singularity,[17] which also follows from the dynamic group-renormalization approach[21] in which the critical exponent of the temperature dependence turns out to be $\varepsilon/19$ in the linear in $\varepsilon = 4 - d$ approximation, where d is the dimensionality of space.

4. An important consequence of the above problem on the spectral composition of scattered light is the appearance of new components as the scattering multiplicity is increased. The experimental detection of these components requires careful investigation of the spectra in turbid media (in particular, near the critical point). An important question then is the relationship between intensities of different multiplicity and between the intensities of the fine-structure components in the doubly-scattered spectrum. The ratio of the integrated doubly scattered intensity to the singly scattered intensity can readily be estimated from the formula $I_2/I_1 \approx hL/2$, where h is the extinction coefficient and L is a typical linear size of the scattering volume. Thus, in opalescence experiments near the critical vapor-forming state, $h \approx 0.5 - 1.0$ cm^{-1} for $\tau \approx 10^{-4}$ on the critical isochore. Hence, for cells with $L \approx 1$ cm, the contribution of double scattering is very important and reaches some tens of percent.[22] Naturally, further approach to the critical point, which leads to an increase in h, may ensure that double scattering will predominate over single scattering when L is large enough. At the same time, there may also be an increase in the contribution of scattered components of higher multiplicity to the overall intensity of multiple scattering.[2)] Although this situation is possible in principle, it is not, as a rule realized in ordinary experiments because the approach to the critical point along the temperature ($\tau_{min} \gtrsim 10^{-5}$) and field ($z_{min} \gtrsim 10^{-1}$ cm) axes is restricted and because of the relatively small size of the cell $L \lesssim 1$ cm.

The ratios of the integrated intensities of the different fine-structure components of pure doubly-scattered spectrum are given by the following expressions:

$$\frac{J_{(L_0)}}{2(J_{(L_1)}+J_{(L_2)})} \approx \int_{-\infty}^{\infty} (L_0)\,d\Omega_2 \Big/ 2\int_{-\infty}^{\infty} ((L_1)+(L_2))\,d\Omega_2 \sim \tau^{-(\gamma-\alpha)}, \quad (39a)$$

$$\frac{J_{(L_0)}}{2J_{(L_1)}} \approx \int_{-\infty}^{\infty} (L_0)\,d\Omega_2 \Big/ 2\int_{-\infty}^{\infty} (L_1)\,d\Omega_2 \sim \tau^{-(\gamma-\alpha)},$$

$$\frac{J_{(L_1)}}{J_{(L_2)}} \approx \int_{-\infty}^{\infty} (L_1)\,d\Omega_2 \Big/ \int_{-\infty}^{\infty} (L_2)\,d\Omega_2 \sim \tau^{-(\gamma-\alpha)}, \quad (39b)$$

$$\frac{J_{(L_0)}}{2J_{(L_2)}} \approx \int_{-\infty}^{\infty} (L_0)\,d\Omega_2 \Big/ 2\int_{-\infty}^{\infty} (L_2)\,d\Omega_2 \sim \tau^{-2(\gamma-\alpha)}. \quad (39c)$$

The above temperature dependences occur in the neighborhood of the critical isochore ($|\tau|^{\beta\delta} \gg z$) as $k \to 0$. As the critical point is approached in the region where the nonlocal character of the fluctuations becomes more and more important, the exponents in the ratios of components of pure doubly-scattered radiation (39) decrease and tend to zero at the critical point, so that the expressions in (18) and (39) become finite. In accordance with (39), a considerable fraction of the intensity of doubly scattered radiation belongs to the unshifted component (L_0). Additional components are, therefore, expected in those materials for which $I_c/2I_{MB} < 10$ in the singly scattered spectrum. This condition is satisfied near the critical state for phase separation of certain mixtures.[33] However, the very low values of the extinction coefficient ($h \lesssim 0.02$ cm^{-1} for $\tau \approx 10^{-5}$) mean that, if the components $\pm L_2$ are to be observed in such systems, the scattering volumes must have sufficiently large linear dimensions ($L \approx 100$ cm).

The observed fine-structure lines in the multiply-scattered spectrum are difficult to interpret because some of the spectrum components of different multiplicity may overlap and the resolution becomes a problem. Thus, the Mandel'shtam-Brillouin components of the singly-scattered spectrum at certain definite angles of observation ϑ coincide either with the $(\pm L_1)$ components at $\vartheta_1 = \pi/2$ or the components $(\pm L_2)$ at $\vartheta_2 = \cos^{-1}(-2/3)$ in the spectrum of pure doubly-scattered radiation. It is clear that, by varying the angle of observation, it is possible to ensure that the condition for the resolution of the components, namely, $\| \Omega_{c,m} | - \Omega_{c,m+1} \| > \Gamma_m$, Γ_{m+1} is satisfied. Here, it is important to remember that, in accordance with (19a) and (37), the pure acoustic modes M^+, M^{++} and M^-, M^{--} of the spectra of singly and doubly scattered radiation shift in the opposite directions with departure from the angle ϑ_2. When the components $\pm L_2$ are resolved, the ratios given by (39) can be used to subtract the contribution of doubly-scattered radiation to the resultant integrated intensity and thus determine the contribution of single scattering. This seems to be the correct way of taking into account the contribution of higher-order scattering effects when the magnitude of single scattering is estimated.

We note that double scattering is more conveniently observed in a depolarized spectrum from which singly scattered radiation is completely absent.

The kinetic properties of the scattering medium can also be investigated by examining the doubly-scattered spectra. All the information on these properties is contained in the side components $\pm L_1$ and $\pm L_2$, so that there is no need to analyze the degenerate central component L_0 in this case.

Experimental verification of (36) would be of considerable interest for the above theory of multiple scattering. This could be done by separating doubly-scat-

tered radiation from the resultant spectral intensity. This would require independent measurements of the half-widths $\Gamma^{(2)}$ and $\Gamma^{(3)}$ of side components and the half-width $\Gamma^{(1)}$ of the double thermal mode M^{00}. This chould be achieved by subtracting twice the spectral function for the $\pm L_2$ component from the spectral function for the component (L_0) [see (32)].

Investigation of multiply-scattered spectra in spatially inhomogeneous media near the critical point requires analysis of the experimental information obtained for locally isotropic layers.[9,1] The intensity of scattered radiation differentiated with respect to height is then conveniently observed at $\vartheta = \pi/2$. All the measured characteristics of multiply-scattered spectra (entering points, half-widths of components, and their ratios) depend not only on the temperature but also on the field variable z.

Studies of multiple scattering of radiation (integrated intensity and the spectral composition) are a promising and essential way of obtaining information on multiparticle space-time correlations and the associated equilibrium and kinetic properties of condensed media.

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[1)]Here and henceforth, the second figure represents the number of the expression taken from[1], and the notation used there is also employed here.

[2)]In general, the spectrum of pure i-fold scattering contains contributions of 3^i modes, of which one is i-fold thermal, $i(i-1)+2$ is i-fold acoustic, and the rest are mixed. The intensity of pure i-fold scattering is $I_i \sim L^{i+2}$ and the intensity of the interference effects, determined by correlators of order $i+j (j<1)$ is proportional to L^{2i-j+2}.

[1]E. L. Lakoza and A. V. Chalyĭ, Zh. Eksp. Teor. Fiz. 67, 1050 (1974) [Sov. Phys. JETP 40, 521 (1975)].
[2]D. W. Oxtoby and W. M. Gelbert, Phys. Rev. A 10, 738 (1974); J. Chem. Phys. 60, 3359 (1974).
[3]V. L. Kuz'min, Opt. Spektrosk. 39, 546 (1975) [Opt. Spectrosc. (USSR) 39, 306 (1975)].
[4]L. V. Adzhemyan, L. Ts. Adzhemyan, L. N. Zubkov, and V. P. Romanov, Pis'ma Zh. Eksp. Teor. Fiz. 22, 11 (1975) [JETP Lett. 22, 5 (1975)].
[5]N. J. Trappeniers, A. C. Michels, and R. H. Huijser, Chem. Phys. Lett. 34, 192 (1975).
[6]D. Beysens, A. Bourgou, and H. Charlin, Phys. Lett. A 53, 126 (1975).
[7]A. Z. Patashinskiĭ and V. L. Pokrovskiĭ, Fluktuatsionnaya teoriya fazovykh perekhodov (Fluctuation Theory of Phase Transitions), Nauka, 1975.
[8]A. V. Chalyĭ and A. D. Alekhin, Zh. Eksp. Teor. Fiz. 59, 337 (1970) [Sov. Phys. JETP 32, 181 (1971)]; N. L. Tsyganov and A. V. Chalyĭ, Zh. Eksp. Teor. Fiz. 61, 1605 (1971) [Sov. Phys. JETP 34, 856 (1972)].
[9]E. L. Lakoza, V. M. Sysoev, and A. V. Chalyĭ, Zh. Eksp. Teor. Fiz. 65, 605 (1973) [Sov. Phys. JETP 38, 298 (1974)].
[10]B. N. Miller and O. V. Splittorff, J. Opt. Soc. Am. 62, 1291 (1972); Phys. Rev. A 9, 550 (1974).
[11]H. K. Leung and B. N. Miller, Phys. Rev. A 12, 2162 (1975).
[12]A. K. Atokhodzhaev, L. M. Kashaeva, L. M. Sabirov, V. S. Starunov, T. M. Utarova, and I. L. Fabelinskiĭ, Pis'ma Zh. Eksp. Teor. Fiz. 17, 95 (1973) [JETP Lett. 17, 65 (1973)].
[13]A. F. Andreev, Pis'ma Zh. Eksp. Teor. Fiz. 19, 713 (1974) [JETP Lett. 19, 368 (1974)].
[14]A. D. Alekhin, A. Z. Golik, N. P. Krupskiĭ, A. V. Chalyĭ, and Yu. I. Shimanskiĭ, in: "Fizika zhidkogo sostoyaniya" (Physics of the Liquid State), Vol. 1, Kharkov State University, 1973, p. 65.
[15]V. M. Sysoev and A. V. Chalyi. Teor. Mat. Fiz. 18, 410 (1974); 26, 126 (1976).
[16]I. L. Fabelinskiĭ, Molekulyarnoe rasseyanie sveta (Molecular Scattering of Light), Nauka, 1965.
[17]H. E. Stanley, Introduction to Phase Transitions and Critical Phenomena, Oxford University Press, 1971 (Russ. Transl., Mir, 1975).
[18]J. L. Lebowitz and J. K. Percus, Phys. Rev. 122, 1675 (1961).
[19]V. L. Kuz'min, F. M. Kuni, and A. I. Rusanov, Zh. Strukt. Khim. 11, 983 (1970).
[20]N. P. Malomuzh, V. P. Oleĭnik, and I. Z. Fisher, Zh. Eksp. Teor. Fiz. 63, 2336 (1972) [Sov. Phys. JETP 36, 1233 (1972)].
[21]B. J. Halperin, P. C. Hohenberg, and E. D. Siggia, Phys. Rev. Lett. 32, 1289 (1974).
[22]A. D. Alekhin and N. P. Krupskii, Sb. "Fizika zhidkogo sostayaniya" (in: Physics of the Liquid State), Vol. 3, Kharkov State University, 1975, p. 48.
[23]M. A. Anisimov, A. M. Evtyushenkov, Yu. F. Kiyachenko, and I. K. Yudin, Pis'ma Zh. Eksp. Teor. Fiz. 20, 378 (1974) [JETP Lett. 20, 170 (1971)].

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