Multiple scattering of light in an inhomogeneous medium near the critical point

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The general solution is considered of the problem of arbitrary order scattering, near the critical point, of electromagnetic waves in a medium, whose anisotropy is caused by an external field. The general results of the electrodynamic calculation are applied to the study of single and double scattering and the resultant "interference" effects. The singularities of these effects are analyzed by using the results of the scale transformation theory and the expressions for the correlation functions of the scalar order parameter of an anisotropic medium. Some possibilities of an experimental study of critical opalescence in the double scattering approximation are discussed.

The construction of the generally accepted theories of critical opalescence (the anomalous scattering of light, x-rays and neutrons near phase transition points of first order and critical points) takes only single scattering of the radiation into account.[1,2] However, as the critical state is approached, correct description of the phenomenon of critical opalescence requires consideration of higher-order scattering effects.

Near the liquid-vapor critical point, it is necessary to take into account an additional important fact connected with the lowering of the symmetry (disturbance of isotropy) of the medium under the action of the external field in the experimental situation usually involved. This circumstance has been considered in an investigation of single scattering of electromagnetic waves [3] and the structure of pair correlation functions of the fluctuations of a scalar order parameter [4] near the critical point.

In the present study, the results that were obtained previously have been consistently applied to the description of scattering effects of higher order near the critical point in an optically inhomogeneous medium rendered anisotropic by the external field.

CALCULATION OF i–FOLD SCATTERING

The process of propagation of electromagnetic waves in an optically inhomogeneous medium with $\epsilon = \epsilon(\mathbf{r})$, $\mu = 1$ is described by the equation

$$\Delta\mathbf{E}+k_0^2\varepsilon\mathbf{E}-\nabla(\nabla\mathbf{E})=0,$$

from which follows a closed system of coupled differential equations for the fields $\mathbf{E}_i$ of different orders of scattering:

$$\Delta\mathbf{E}_0+k_0^2\varepsilon_0\mathbf{E}_0-\nabla(\nabla\mathbf{E}_0)=0; \tag{1}$$

$$\Delta\mathbf{E}_i+k_0^2\varepsilon_0\mathbf{E}_i=\mathbf{\Phi}_{i-1}-\nabla(\mathbf{E}_i\nabla\ln\varepsilon_0),\quad \mathbf{\Phi}_i=-k_0^2\varepsilon'\mathbf{E}_i-\nabla\left(\frac{1}{\varepsilon_0}\mathbf{E}_i\nabla\varepsilon'\right). \tag{2}$$

Here $k_0 = 2\pi/\lambda$, $\epsilon = \epsilon_0 + \epsilon'$, $\epsilon_0(\mathbf{r})$ and $\epsilon'(\mathbf{r})$ are the macroscopic and fluctuating parts of the dielectric permittivity, $i = 1, 2, \ldots$.

We assume that the scattering takes place in a plane-parallel layer $-L_Z \le z \le L_Z$ that is inhomogeneous in z, with normal incidence of the exciting wave E_0 on the boundary $z = -L_Z$. We shall also assume the inhomogeneity created by the external field to be macroscopic in the sense that its characteristic dimension $R_0 \approx |\nabla \ln \epsilon_0|^{-1}$ satisfies the inequality $R_0 \gg R_C$, where R_C is the correlation radius of the density fluctuations.

The field of the exciting wave in the volume of the investigated layer can be found by the WKB method, the criterion of applicability of which is the condition

$$|\nabla\varepsilon_0|/k_0\ll 1 \tag{3}$$

(the equivalent criterion $\tau = (T-T_C)/T_C \gg 10^{-3}$ was obtained ealier [3]). The field is given by

$$\mathbf{E}_0(z)=\frac{1}{\varepsilon_0^{1/4}(z)}\left\{\mathbf{A}\exp\left[ik_0\int_{-L_z}^{z}\varepsilon_0^{1/2}(z')dz'\right]+\mathbf{B}\exp\left[-ik_0\int_{-L_z}^{z}\varepsilon_0^{1/2}(z')dz'\right]\right\}. \tag{4}$$

To find the ith-order scattered wave, we write down the following integral equation, which is equivalent to (2):

$$\mathbf{E}_i(r)=\int G(\mathbf{r},\mathbf{r}')\{\mathbf{\Phi}_{i-1}(\mathbf{r}')-\nabla'(\mathbf{E}_i(\mathbf{r}')\nabla'\ln\varepsilon_0(z'))\}d\mathbf{r}'; \tag{5}$$

here $G(\mathbf{r}, \mathbf{r}')$ is the Green's function of the operator of the left side of (2), which, in the smooth-inhomogeneity approximation [1)], has the form

$$G_0(\mathbf{r},\mathbf{r}')=-\frac{1}{4\pi|\mathbf{r}-\mathbf{r}'|}\exp[ik_0\varepsilon_0^{1/2}(z')|\mathbf{r}-\mathbf{r}'|]. \tag{6}$$

After the obvious transformations in (5), under the wave-zone condition

$$k_0\varepsilon_0^{1/2}(z')|\mathbf{r}-\mathbf{r}'|\gg 1 \tag{7}$$

we get for $\mathbf{E}_i$, with account of (3):

$$\mathbf{E}_i(\mathbf{r})=-k_0^2\int\varepsilon'(\mathbf{r}_i)G_0(\mathbf{r},\mathbf{r}_i)[\mathbf{n}_i\times[\mathbf{E}_{i-1}(\mathbf{r}_i)\times\mathbf{n}_i]]d\mathbf{r}_i \tag{8}$$

and, similarly,

$$\mathbf{H}_i(\mathbf{r})=\frac{1}{ik_0}[\nabla\times\mathbf{E}_i]=-k_0^2\int\varepsilon'(\mathbf{r}_i)\varepsilon_0^{1/2}(z_i)G_0(\mathbf{r},\mathbf{r}_i)[\mathbf{n}_i\times\mathbf{E}_{i-1}(\mathbf{r}_i)]d\mathbf{r}_i, \tag{9}$$

where $\mathbf{n}_i$ is a unit vector directed from the point of ith-order scattering to the point of observation.

In what follows, it is convenient to define the Umov-Poynting vector, which has direct experimental interest:

$$\langle\mathbf{S}_{ii}\rangle=\frac{c}{8\pi}\mathrm{Re}\left\{\langle[\mathbf{E}_i\times\mathbf{H}_i^*]\rangle+\sum_{j=0}^{i-1}(\langle[\mathbf{E}_i\times\mathbf{H}_j^*]\rangle+\langle[\mathbf{E}_j\times\mathbf{H}_i^*]\rangle)\right\}. \tag{10}$$

The first term in (10) represents "pure" ith-order scattering, and the rest the so-called "interference" effects. In the general case, the accuracy of calculations of the terms in (10) depends on the explicit form of the multipoint correlators

$$g_{l+m}(\mathbf{r}_1,\ldots,\mathbf{r}_l,\mathbf{r}_1',\ldots,\mathbf{r}_m')=\left\langle\prod_{k=1}^{l}\Delta\rho(\mathbf{r}_k)\prod_{n=1}^{m}\Delta\rho(\mathbf{r}_n')\right\rangle \tag{11}$$

$(l + m \le 2i)$, where the angle brackets denote averaging over the locally equilibrium distribution function (isothermal case).[5] The structure of (11) for an arbitrary configuration is unknown at the present time near the critical point. However, the wave-zone condition (7), applied above in the electrodynamic calculation, allows us to use the asymptotic expressions obtained in a number of studies [6-10] for the correlation function of the Bogolyubov-Born-Green-Kirkwood-Yvon (BBGKY) theory and the correlation function of Ursell-Mayer, which corresponds to the removal of one group of particles from another to a large distance.

Unfortunately, the temperature criterion of the WKB method does not allow us to apply the results of Polyakov,[11] who found an explicit form of the 3-point correlators on the basis of the hypothesis of conformal invariance of the critical fluctuations and imposed limitations on the structure of the 4-point correlators at the critical point. The general formulas of this section will be used below for calculation of single and double scattering with their interference effects in an anisotropic medium close to the critical point.

SINGLE-SCATTERING APPROXIMATION

We shall assume that a study of the scattering ability of a material in a near-critical state can be carried out in the single-scattering approximation. In the case of pure single scattering, we obtain for $\langle S_{11}^0\rangle$, using the results of [9,10] and assuming that the linear dimensions of the scattering volume are much smaller than the distance from it to the point of observation ($2L_Z \ll L$, $\mathbf{n}_1' \approx \mathbf{n}_1$),

$$\langle S_{11}^0\rangle = \frac{c}{8\pi}\left(\frac{k_0^2}{4\pi}\right)^2\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_T^2\frac{\mathbf{n}_1(1-(\mathbf{n}_1\mathbf{m}_0)^2]}{L^2}\cdot \tag{12}$$

$$\times \operatorname{Re}\int g_2(\mathbf{r}_1,\mathbf{r}_1')\,\varepsilon_0^{1/2}(z_1')\exp\{ik_0\varepsilon_0^{1/2}(z_1')\,(\mathbf{n}_1(\mathbf{r}_1-\mathbf{r}_1'))\}\mathbf{E}_0(z_1)\mathbf{E}_0^*(z_1')\,d\mathbf{r}_1\,d\mathbf{r}_1',$$

where $\mathbf{m}_0$ is the polarization vector of the exciting wave and L is the distance from the scattering volume to the observation point.

Transformations in (12) similar to those used in [3] give the following expression for the intensity of pure single scattering $I_1 = \mathbf{n}_1\langle S_{11}^0\rangle$:

$$I_1 = I_0\frac{2\pi\sigma}{L^2}(1-(\mathbf{n}_1\mathbf{m}_0)^2)\int_{-L_z}^{L_z}\frac{\alpha(z,\tau)}{w_0(z,\tau,\mathbf{n}_1,\mathbf{n}_0)}dz, \tag{13}$$

where $I_0 = c|A|^2/8\pi$ is the intensity of the incident light beam of cross section σ,

$$\alpha(z,\tau) = \frac{\pi}{2\lambda^4}\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_T^2 k_B T\beta_T(z,\tau),$$

$$w_0(z,\tau,\mathbf{n}_1,\mathbf{n}_0) = 1+\delta(z,\tau)\,(1-(\mathbf{n}_1\mathbf{n}_0)),$$

$$\delta(z,\tau) = 8\pi^2\lambda^{-2}f^*(z,\tau)\beta_T(z\ \tau),\qquad f^*(z,\tau) = p_c b\varepsilon_0(z,\tau),$$

$\beta_T(z,\tau) = [p_c b\kappa_{\text{eff}}^2(z,\tau)]^{-1}$ is the local value of the isothermal compressibility, and $\mathbf{n}_0 = \{0, 0, \mathbf{e}_3\}$ is the direction of the incident exciting wave. In obtaining (13), we did not take into account effects connected with the reverse wave in (4) 2) and the expression (from [4]) used for $g_2(\mathbf{r}_1, \mathbf{r}_1')$ was of the form

$$g_2(\mathbf{r}_1,\mathbf{r}_1') = \frac{k_B T}{4\pi b p_c|\mathbf{r}_1-\mathbf{r}_1'|}\exp[-\varkappa_{\text{eff}}(z_1'\tau)\,|\mathbf{r}_1-\mathbf{r}_1'|]. \tag{14}$$

Here

$$\varkappa_{\text{eff}}(z,\tau) = [b^{-1}\tau^{-\gamma}dG(y(z,\tau))/dy]^{1/2}$$

is the inverse correlation radius of the density fluctuations, $G(y)$ is the scale function of the equation of state of similarity theory,[12-15] and b is the coefficient in front of the gradient term in the expression for the fluctuation part of the free energy;[4] $\gamma \approx 1.25$ is the critical index of $\beta_T(\tau)$.

In studies of single scattering in an inhomogeneous medium, the experimental information should be taken from a layer in which the properties of the system differ insignificantly in the different directions. The differential intensity of radiation singly scattered in such a layer of "local isotropy" (the corresponding criteria are given in [3]) has the form

$$\frac{dI_1}{dz} = I_0\frac{2\pi\sigma}{L^2}\frac{\alpha(z,\tau)}{w_0(z,\tau,\mathbf{n}_1,\mathbf{n}_0)}(1-(\mathbf{n}_1\mathbf{m}_0)^2). \tag{15}$$

on the basis of (13). In the case of a homogeneous system,

$$G_y' \to A,\quad \varepsilon_0(z,\tau)\to\varepsilon_c,\quad \varkappa_{\text{eff}}^2(z,\tau)\to A\tau^\gamma/b,$$

$$\beta_T^{-1}(z,\tau)\to p_c A\tau^\gamma$$

and Eq. (13) becomes the well-known formula for the intensity of single scattering of the Ornstein-Zernike theory.[1,2,16]

The second and third terms in (10), which describe interference effects, vanish in the model of Gaussian fluctuations. However, in a model that takes into account the small departure from a purely Gaussian distribution, these interference effects turn out to be different from zero. In particular, the use of the quasi-Gaussian macroscopic distribution function of [17,18] shows that the interference effects of first order appear only in the direction of propagation of the exciting wave: $I_{1/2} \sim g_1(\mathbf{n}_1 = \mathbf{n}_0)$. A thermodynamic estimate of the zeroth Fourier-component of single-point correlation function of the density fluctuations in this model gives

$$g_1(\mathbf{n}_1=\mathbf{n}_0)\sim(\partial^2\mu/\partial\rho^2)_T(\partial\mu/\partial\rho)_T^{-2}.$$

This result gives reason to doubt the advisibility of using the quasiclassical-fluctuation model in what follows to calculate correlation functions of odd orders, since interference effects of first order should be absent, being determined by the linear fluctuation, which is identically equal to zero.

THE DOUBLE-SCATTERING APPROXIMATION

We first carry out the double-scattering calculation in the pure scattering approximation, which is connected with the first term $\langle S_{22}^0\rangle$ in (10) for $i = 2$. Using (9), (10), we get the following expression for the intensity of pure double scattering $I_2 = (\mathbf{n}_2\langle S_{22}^0\rangle)$ in a volume V whose linear dimensions are much less than the distance from it to the point of observation ($|\mathbf{r}-\mathbf{r}_2| \approx |\mathbf{r}-\mathbf{r}_2'| \approx L$, $\mathbf{n}_2' \approx \mathbf{n}_2$):

$$\begin{aligned}I_2 = {}&\frac{c}{8\pi}\left(\frac{k_0^2}{4\pi}\right)^4\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_T^4\operatorname{Re}\int g_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_1',\mathbf{r}_2')\,\varepsilon_0^{1/2}(z_1')\,\varepsilon_0^{1/2}(z_2')\\ &\times\mathbf{E}_0(z_1)\mathbf{E}_0^*(z_1')\,|\mathbf{r}_2-\mathbf{r}_1|^{-1}|\mathbf{r}_2'-\mathbf{r}_1'|^{-1}\exp\{ik_0[\varepsilon_0^{1/2}(z_2)\,|\mathbf{r}-\mathbf{r}_2|\\ &+\varepsilon_0^{1/2}(z_1)\,|\mathbf{r}_2-\mathbf{r}_1|-\varepsilon_0^{1/2}(z_2')\,|\mathbf{r}-\mathbf{r}_2'|-\varepsilon_0^{1/2}(z_1')\,|\mathbf{r}_2'-\mathbf{r}_1'|]\}\\ &\times\{|\mathbf{m}_0|^2-(\mathbf{n}_2\mathbf{m}_0)^2-(\tilde{\mathbf{n}}_1\mathbf{m}_0)^2-(\tilde{\mathbf{n}}_1'\mathbf{m}_0)^2+(\tilde{\mathbf{n}}_1\mathbf{m}_0)\,(\tilde{\mathbf{n}}_1'\mathbf{m}_0)\,(\mathbf{n}_1\tilde{\mathbf{n}}_1')\\ &+(\tilde{\mathbf{n}}_1\mathbf{m}_0)\,(\tilde{\mathbf{n}}_1\mathbf{n}_2)\,(\mathbf{n}_2\mathbf{m}_0)+(\tilde{\mathbf{n}}_1\mathbf{m}_0)\,(\tilde{\mathbf{n}}_1'\mathbf{n}_2)\,(\mathbf{n}_2\mathbf{m}_0)-\\ &-(\tilde{\mathbf{n}}_1\mathbf{m}_0)\,(\tilde{\mathbf{n}}_1'\mathbf{m}_0)\,(\tilde{\mathbf{n}}_1\mathbf{n}_2)\,(\tilde{\mathbf{n}}_1'\mathbf{n}_2)\}d\mathbf{r}_1\,d\mathbf{r}_2\,d\mathbf{r}_1'\,d\mathbf{r}_2'.\end{aligned} \tag{16}$$

where $\mathbf{n}_1$ and $\mathbf{n}_1'$ are unit vectors that indicate the direction of the first scattering act inside the volume V over which the integration in (16) is carried out.

We consider in more detail a configuration of four fluctuations whose contribution is dominant within the framework of the assumptions made previously, namely: two pairs of fluctuations, with distances between the pairs $\mathbf{R}_1=\mathbf{r}_1-\mathbf{r}_1'$ and $\mathbf{R}_2=\mathbf{r}_2-\mathbf{r}_2'$ of the order of the correlation radius R_C, are separated by a distance $|\mathbf{r}_2-\mathbf{r}_1|\approx|\mathbf{r}_2'-\mathbf{r}_1'|$ that satisfies the wave-zone condition (7). For this configuration, we obtain the asymptotic structure of the 4-point correlation function of the density fluctuations $g_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_1',\mathbf{r}_2')$ by using the results of Lebowitz and Percus:[6]

$$g_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_1',\mathbf{r}_2')=g_2(\mathbf{r}_1,\mathbf{r}_1')g_2(\mathbf{r}_2,\mathbf{r}_2')-\frac{k_BT\bar\rho^2\beta_T}{V}\frac{\partial g_2(\mathbf{r}_1,\mathbf{r}_1')}{\partial\bar\rho}\frac{\partial g_2(\mathbf{r}_2,\mathbf{r}_2')}{\partial\bar\rho}$$
$$+\frac{k_BT\bar\rho^2\beta_T}{V}\frac{\partial}{\partial\bar\rho}(F_1(\mathbf{r}_1)F_1(\mathbf{r}_1'))\frac{\partial}{\partial\bar\rho}(F_1(\mathbf{r}_2)F_1(\mathbf{r}_2'))$$
$$+\hat P[F_1(\mathbf{r}_1)F_1(\mathbf{r}_2)g_2(\mathbf{r}_1',\mathbf{r}_2')],\qquad(17)$$
$$\bar\rho\beta_T=\frac{1}{V}\left(\frac{\partial V}{\partial\mu}\right)_T\sim\tau^{-\gamma},$$

where $\hat P$ is the operator of the sum of cyclic permutations of the points, $F_1(\mathbf{r})$ is the one-component correlation function of BBGKY theory, $\bar\rho=N/V$ is the mean density of the system, and N is the number of particles in the volume V.

The first term in (17), corresponding to the Gaussian fluctuation model, makes the basic contribution to I_2, which has the following form if we consider only the direct exciting wave in (4) and expression (14) for $g_2(\mathbf{r},\mathbf{r}')$, and carry out the integration in (16) over $\mathbf{R}_1$ and $\mathbf{R}_2$ in a volume $V\gg R_C^3$,

$$I_2^0=I_0\frac{4\pi^2}{L^2}\int\frac{d\mathbf{r}_1'\,d\mathbf{r}_2'}{|\mathbf{r}_2'-\mathbf{r}_1'|^2}\frac{w_1(\tilde{\mathbf{n}}_1',\mathbf{m}_0,\mathbf{n}_2)}{w_0(z_1',\tau,\tilde{\mathbf{n}}_1',\mathbf{n}_0)}$$
$$\times\frac{\alpha(z_1',\tau)\alpha(z_2',\tau)\varepsilon_0^{1/2}(z_2')}{1+{}^1/_2\delta(z_2',\tau)\varepsilon_0^{-1}(z_2')|\varepsilon_0^{1/2}(z_1')\tilde{\mathbf{n}}_1'-\varepsilon_0^{1/2}(z_2')\mathbf{n}_2|^2},\qquad(18)$$
$$w_1(\tilde{\mathbf{n}}_1',\mathbf{m}_0,\mathbf{n}_2)=1-(\mathbf{n}_2\mathbf{m}_0)^2+(\tilde{\mathbf{n}}_1'\mathbf{m}_0)^2-(\tilde{\mathbf{n}}_1'\mathbf{m}_0)^2(\tilde{\mathbf{n}}_1'\mathbf{n}_2)^2$$
$$+2(\tilde{\mathbf{n}}_1'\mathbf{m}_0)(\tilde{\mathbf{n}}_1'\mathbf{n}_2)(\mathbf{n}_2\mathbf{m}_0).$$

The results of calculation of the contributions of the remaining terms of (17) to the total intensity I_2 are given in the Appendix. As the estimates show, in the temperature range $\tau\gg10^{-8}$ determined by the criterion of applicability of the WKB method,[3] the values of these contributions are found to be negligibly small in comparison with (18).

For analysis of the experimental information obtained for scattering on layers possessing local isotropy, as in the single-scattering approximation, it is convenient to use the differential intensity dI_2^0/dz, for which we have from (18)

$$\frac{dI_2^0}{dz}=I_0\frac{4\pi^2v}{L^2}\int_\Omega\frac{\alpha^2(z,\tau)\varepsilon_0^{1/2}(z)w_1(\tilde{\mathbf{n}}_1,\mathbf{m}_0,\mathbf{n}_2)}{w_0(z,\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_0)w_0(z,\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_2)}d\Omega,\qquad(19)$$

where v is the volume of the layer of local isotropy. Here the integration over the solid angle sums over the directions $\tilde{\mathbf{n}}_1$ the single scatterings in the given layer of local isotropicity that are responsible for the pure double scattering that escapes to the receiver.

In a homogeneous layer, we get the following from (18) for I_2^0 in the volume V:

$$I_2^0=I_0\frac{8\pi^2VL_z\sqrt{\varepsilon_c}}{L^2}\alpha^2(\tau)i_0(\tau,\mathbf{m}_0,\mathbf{n}_0,\mathbf{n}_2),$$
$$i_0(\tau,\mathbf{m}_0,\mathbf{n}_0,\mathbf{n}_2)=\int_\Omega\frac{w_1(\tilde{\mathbf{n}}_1,\mathbf{m}_0,\mathbf{n}_2)}{w_0(\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_0)w_0(\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_2)}d\Omega.\qquad(20)$$

Far from the critical point, when the parameter $\delta=0$ (the Rayleigh-Einstein approximation), the intensity of pure double scattering for natural incident light is described by the formula

$$I_2^{\mathrm{RE}}(\vartheta)=I_0\frac{8\pi^2VL_z\varepsilon_c^{1/2}}{15L^2}\alpha^2(\tau)(11+7\cos^2\vartheta).\qquad(21)$$

It is seen from (21) that the scattering function I_2^{RE} is, as is to be expected, a smoother function of the scattering angle ϑ than the function $I_1^{\mathrm{RE}}(\vartheta)\sim1+\cos^2\vartheta$.

With the approach to the critical point, in the range of temperatures in which we can neglect correlation effects, the intensity of double scattering (21) increases in proportion to $\beta_T^2\sim\tau^{-2\gamma}$. It must be noted that neglect of correlation effects results in singularities in the manifestation of the gravitational effect in the liquid-vapor system—singularities which consist of a sharp decrease in the spatial region of the "true" critical state as $\tau\to0$.[19] Outside this region, which is immediately adjacent to the level with maximum density gradient, the system can be regarded as approximately macroscopically homogeneous, and the calculation of double scattering can be made with sufficient accuracy from the formula (21), even for temperatures very close to critical.

We consider the formula (20) for $\mathbf{n}_2\approx\mathbf{n}_0$ (forward scattering). In this case, the integral is easily calculated and we get for I_2^0 (approximately)

$$I_2^0(\mathbf{n}_2\approx\mathbf{n}_0)=I_0\frac{16\pi^3VL_z}{L^2}\frac{\alpha^2(\tau)}{\delta^2(\tau)}\varepsilon_c^{1/2}\left[\delta(\tau)-\frac{1}{2}\ln\delta(\tau)\right].\qquad(22)$$

It is then seen that $I_2^0(\mathbf{n}_2\approx\mathbf{n}_0)$ contains two singular contributions proportional to β_T and $\ln\beta_T$. This result is in qualitative agreement with the conclusion of [10], which considered a configuration of two pairs of points compressed to molecular separation distances within each pair ($|\mathbf{r}_1-\mathbf{r}_1'|\approx|\mathbf{r}_2-\mathbf{r}_2'|\approx a_0$) and separated by a distance $|\mathbf{r}_1-\mathbf{r}_2|\approx|\mathbf{r}_1'-\mathbf{r}_2'|\approx R_C$. As is shown in [10], the 4-point correlation function of such a configuration for a homogeneous liquid near the critical point gives two contributions: a slowly decreasing one, connected with the singular behavior of the susceptibility β_T, and a more rapidly decaying one, connected with the singular behavior of the specific heat c_V. The singularity of $c_V\sim\tau^{-\alpha}$ ($\alpha\approx0.12$) is similar in character to the singularity of $\ln\beta_T\sim\ln\tau$.

In the immediate neighborhood of the critical point, the most important configuration is that for which all the distances between the fluctuations turn out to be of the order of R_C. An estimate of the principal contribution of this configuration to the expression for I_2 evidently requires the use of considerations of conformal invariance of the critical fluctuations.[11]

We now proceed to the study of second-order interference effects in (10) for $i=2$. The terms determined by the second-order correlation function turn out to be negligible on the ground that they contain $g_2(\mathbf{r}_1,\mathbf{r}_2)$, in which the relative separation of the fluctuations $|\mathbf{r}_1-\mathbf{r}_2|\gg\lambda$, in correspondence with the condition of the wave zone (7), since the important distances in $g_2(\mathbf{r}_1,\mathbf{r}_2)$ under normal experimental conditions are $|\mathbf{r}_1-\mathbf{r}_2|\approx R_C<\lambda$. Then, for the second-order interference terms

$$\langle\mathbf{S}_{1/2}\rangle=\frac{c}{8\pi}\mathrm{Re}\{\langle[\mathbf{E}_2\times\mathbf{H}_1^*]\rangle+\langle[\mathbf{E}_1\times\mathbf{H}_2^*]\rangle\}\qquad(23)$$

upon satisfaction of the wave-zone condition (7) between the points $\mathbf{r}_1$ and $\mathbf{r}_2$ at which successive scattering acts occur in the volume V, the principal contribution to (23) is made by the configuration in which the strongly correlating fluctuations are at distances $|\mathbf{r}_1-\mathbf{r}_1'|\approx R_C$. For such a configuration, the use of the asymptotic expressions of [6] for $g_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_1')$ and Eq. (14) leads to the following expression for the differential intensity:

$$\frac{dI_{3/2}}{dz}=\frac{d}{dz}(\mathbf{n}_2\langle\mathbf{S}_{3/2}\rangle)=I_0B(z,\tau)\alpha(z,\tau)$$
$$\times\left\{\frac{\partial\ln b}{\partial\bar\rho}J(a)\Big|_{a=1}-\frac{\partial\ln\varkappa^2_{\rm eff}}{\partial\bar\rho}\frac{\partial J(a)}{\partial a}\Big|_{a=1}\right\},$$
$$B(z,\tau)=\frac{\pi^2}{L^2\lambda^2}\left(\rho\frac{\partial\varepsilon}{\partial\bar\rho}\right)\frac{k_BT\bar\rho^2\beta_T}{V}\frac{\partial F_1(z)}{\partial\bar\rho}[1+\varepsilon_0^{-1/2}(z)]vd, \tag{24}$$
$$J(a)=\int\limits_\Omega\frac{(\mathbf{n}_1\mathbf{m}_0)(\mathbf{n}_1\times[\mathbf{m}_0\times\mathbf{n}_2])}{a-1+w_1(z,\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_0)}d\Omega,$$

where d is the thickness of the layer of local isotropy.[3] The solid-angle integrals J(a) and $\partial J(a)/\partial a$ at $a=1$ sum over the directions $\tilde{n}_1$ the single scattering in the volume which is responsible for the interference contribution of second order registered by the receiver.

Analysis of expression (24), like that of the corresponding formula for the homogeneous case, shows that $dI_{3/2}/dz$ turns out to be much less than dI_2/dz, as is seen from the following estimate for the region where one can neglect the correlation effects ($\delta\ll 1$):

$$\frac{dI_{3/2}}{dz}\Big/\frac{dI_2}{dz}\sim\frac{\tau^\nu}{N}.$$

Moreover, $dI_{3/2}/dz=0$ at $\mathbf{n}_2\mathbf{n}_0=1$ and $\mathbf{n}_2\mathbf{n}_0=0$, i.e., second-order interference effects are absent, both in the direction of propagation of the incident light beam and in the perpendicular direction.

DISCUSSION OF RESULTS

The consideration that has been given to the problem of electromagnetic wave propagation in matter near the critical point reveals a strong dependence of the scattering properties on the "field" variable (in the specific case of a gravitational field, on the height measured from the "critical" level with the maximum density gradient). It is at the critical level that the scattering ability of the material is an extremum at fixed τ. Experimental investigations confirming this fact, the results of which are given in [19], show that, for example, at $\tau\approx 10^{-4}$ the intensities of single scattering at the levels $z\approx 0$ and $z\approx 1$ cm differ by two orders of magnitude. A study of the height dependence of the scattering ability near the critical point, based on the use of the gravitational effect, contains valuable information (supplementing the now traditional studies of the temperature dependence, in which an attempt is usually made to eliminate the gravitational effect by any means) on the equation of state in the vicinity of the critical point, on the field critical indices, and so on.

The study of light scattering in an optically inhomogeneous medium, as has been noted, is conveniently carried out in layers of local isotropy. For the geometry considered here, this can be accomplished by observing the scattered radiation at an angle $\pi/2$ with respect to the direction of the incident beam. In this case, for treatment of the experimental data in the approximations of single and double scattering, it is necessary to use Eqs. (15) and (19) for the corresponding differential cross sections, in which one should set $\mathbf{n}_1\cdot\mathbf{n}_0=0$ and $\mathbf{n}_2\cdot\mathbf{n}_0=0$, respectively. The choice of this direction of observation is convenient also for the reason that here the contribution of the second-order interference effects is equal to zero (see (24)).

Interesting experimental possibilities are inherent in study of the scattering ability of matter in an optically inhomogeneous medium in the double-scattering approximation near the critical point with the use of polarization effects. Thus, from Eqs. (15), at $\mathbf{n}_1=\mathbf{m}_0$ and from (19) at $\mathbf{n}_2=\mathbf{m}_0$, it follows that $dI_1^{\parallel}/dz=0$, $dI_2^{\parallel}/dz\neq 0$, while at $\mathbf{n}_1,\mathbf{n}_2\perp\mathbf{m}_0$ and $\mathbf{n}_1,\mathbf{n}_2\perp\mathbf{n}_0$, we have $dI_1^{\perp}/dz\neq 0$ and $dI_2^{\perp}/dz\neq 0$. The quantity $dI_{3/2}/dz$ is equal to zero in both cases. We propose to consider this question in some detail in the future.

We have given a general solution of the problem of scattering of radiation of arbitrary order. The only approximation used in the calculation was the wave-zone condition (7), which selects periodic solutions of Eq. (2) far from the scattering center. In the temperature interval determined by the WKB condition (3), this condition required the use of asymptotic expressions for the correlation functions of the type (17). As a result, it turned out that the character of the singularities is about the same for I_1, $I_{3/2}$ and I_2; it is described by the behavior of the isothermal compressibility. It must be noted that in the immediate vicinity of the critical point, where $R_C\gg\lambda$, condition (7) does not impose any limitations on the structure of the correlation function. Here formulas (8) and (9) are exact. However, their use requires, firstly, knowledge of the structure of the multipoint correlation functions at the critical point and, secondly, solution of Eq. (1) for the exciting wave E_0 under the condition that the derivative $\partial\epsilon_0/\partial z\to\infty$ at the point $z=0$ as $\tau\to 0$.

APPENDIX

The contribution of the correlation departure $g_4-g_2g_2$ from expression (17) to the intensity I_2 is found with the help of the relation

$$\frac{\partial g_2(\mathbf{r}_i,\mathbf{r}_j)}{\partial\bar\rho}=-g_2(\mathbf{r}_i,\mathbf{r}_j)\frac{\partial}{\partial\bar\rho}[\ln b+|\mathbf{r}_i-\mathbf{r}_j|\varkappa_{\rm eff}(z_j,\tau)].$$

For simplicity of presentation below, the results of the calculation are given only in the homogeneous case. The contributions of the second and higher terms from (17) are equal to

$$\frac{I_2^{(1)}}{I_2^0}=\frac{k_BT\bar\rho^2\beta_T}{V}\left\{\left(\frac{\partial\ln b}{\partial\bar\rho}\right)^2+\frac{\partial\ln b}{\partial\bar\rho}\frac{\partial\ln\varkappa^2(\tau)}{\partial\bar\rho}\frac{i_1}{i_0}+\left(\frac{\partial\ln\varkappa^2(\tau)}{\partial\bar\rho}\right)^2\frac{i_2}{i_0}\right\},$$
$$\frac{I_2^{(2)}}{I_2^{(0)}}=\frac{4\pi V(\partial F_1^2/\partial\bar\rho)^2\bar\rho\delta_{\mathbf{n}_2\mathbf{n}_0}}{k_BT\beta_Ti_0}, \tag{A.1}$$
$$\frac{I_2^{(3)}}{I_2^0}=\frac{16\pi VF_1^2[1-(\mathbf{n}_2\mathbf{m}_0)^2]}{k_BT\beta_Ti_0}$$
$$i_1=\int w_1(\tilde{\mathbf{n}}_1,\mathbf{m}_0,\mathbf{n}_2)[w_0^{-2}(\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_0)w_0^{-1}(\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_2)+w_0^{-1}(\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_0)w_0^{-2}(\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_2)]d\Omega,$$
$$i_2=\int w_1(\tilde{\mathbf{n}}_1,\mathbf{m}_0,\mathbf{n}_2)w_0^{-2}(\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_0)w_0^{-2}(\tau,\tilde{\mathbf{n}}_1,\mathbf{n}_2)d\Omega.$$

The integrals i_0, i_1 and i_2 are calculated in a specially chosen orthogonal basis. For small scattering angles ($\mathbf{n}_2\approx\mathbf{n}_0$)

$$i_0=\frac{2\pi}{\delta^3}\left(\delta-\frac{1}{2}\ln\delta\right),\quad i_1=\frac{2\pi}{\delta^3}\left(\delta^2-\frac{1}{2}\delta+\frac{3}{4}\ln\delta\right),$$
$$i_2=\frac{2\pi}{3\delta^3}\left(\delta^2-\frac{1}{4}\delta\right).$$

In the temperature range $\tau \gg 10^{-8}$, the contributions (A.1) to I_2 turn out to be small, as shown by the estimates

$$I_2^{(1)}/I_2^0 \sim \tau^{-\gamma} N^{-1}, \quad I_2^{(2)}/I_2^0 \sim \tau^{\gamma} \delta N^{-1},$$

$$I_2^{(3)}/I_2^0 \sim \tau^{\gamma} \delta^{-1}.$$

Here the maximum value of the parameter $\delta(\tau)$, which corresponds to $\tau \approx 10^{-8}$, using $p_c \approx 10^7$ dyn/cm^2, $\lambda = 5 \times 10^{-5}$ cm, and $f^* = 10^{-9}$ dyn [20], is of the order of $10^2 - 10^3$.

1) To find the exact Green's function G(r, r′), the method developed in [4] for the operator $\hat{L} = -\Delta + k_0^2 \epsilon_0$ can be used.

2) As a consequence of allowance for the backward wave, the single-scattering cross section at the critical point diverges in the direction of a scattering angle of π. [3]

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