Multiple scattering of light in an inhomogeneous medium near the critical point. IV. Relation between scatterings of different orders. The extinction coefficient

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The intensity of radiation multiply scattered in a fluid in the near-critical state is calculated. The extinction coefficient for mulitple scattering is computed, and the conditions under which the concept of scattering multiplicity loses its original meaning are obtained. The theoretical results are illustrated by a numerical computation of the extinction coefficient in the Ornstein-Zernike approximation. The results of the present paper together with those of an earlier calculation of the depolarization coefficient in the critical region allow us to assert the effects of multiple scattering are important when $qR_c \gtrsim 1$ (R_c is the correlation length and q is the wave-vector transfer). This agrees with the results of a number of experimental investigations.

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We need to have a deep understanding of the characteristics of multiple light scattering in the critical region for the following main reasons: 1) multiple light scattering furnishes specific information about the higher-order space-time correlation functions (CF) of different physical quantities; 2) allowance for the effects of multiple scattering is necessary for the correct interpretation of the results of the investigation of the phenomenon of critical opalescence in the single-scattering approximation, for which there is a reliable theory.[1,2]

The first theoretical calculations of double light scattering in a fluid near the critical point were published in 1974. These calculations were based on a consistent electrodynamic calculation of the scattered fields and the use of density-fluctuation correlation functions that are right for the purpose of calculating the depolarization coefficient[3] and the integrated intensity[4] of scattered radiation. Since then there has been accumulated a large amount of experimental and theoretical data on the characteristics of the integrated and spectral intensities of higher-order scatterings (see, for example, Refs. 5–19).

One of the central questions is the question of the relation between different orders of scattering in the critical region. This is the subject of the present paper, which continues the investigations started in our previous papers.[4,12,19]

First we shall derive recursion formulas for the integrated intensities (scattering coefficients) for different scattering orders and find a closed solution to the integral equation for the total multiple-scattering coefficient. Then we shall determine the extinction coefficient for multiple scattering, and discuss the question of the relation between the contributions of successive orders. And, finally, we shall carry out specific calculations and numerical estimates of the extinction coefficient for multiple scattering in the Ornstein–Zernike (OZ) approximation. Here special attention will be given to the analysis of the condition for the convergence of the iteration series in the scattering order and the computation of those values of the parameter qR_c (R_c is the correlation length and q is the wave-vector transfer) at which it becomes impossible to use the original concept of scattering order [in the sense of the Born (Rayleigh) expansion] in the solution of the electrodynamic problem.

1. Let us derive the recursion formula for the intensity of the i-tuple scattering:

$$I_i = \langle J_i \rangle = \frac{c}{8\pi} \langle [\mathbf{E}_i \times \mathbf{H}_i^*] \rangle \mathbf{n}_i, \tag{1}$$

where $\mathbf{n}_i$ is the unit vector specifying the direction of the i-tuple scattering and $\langle \ldots \rangle$ denotes statistical averaging over the local-equilibrium density-fluctuation distribution function in the isothermal case.

The general formulas obtained for the scattered fields $\mathbf{E}_i$ and $\mathbf{H}_i$ in the wave-zone approximation have the following form:

$$\mathbf{E}_i(\mathbf{r}) = k_0^2 \int_V \varepsilon'(\mathbf{r}_i) G_0(\mathbf{r}, \mathbf{r}_i) \mathbf{m}_i E_{i-1}(\mathbf{r}_i) d\mathbf{r}_i, \tag{2}$$

$$\mathbf{H}_i(\mathbf{r}) = k_0^2 \int_V \varepsilon'(\mathbf{r}_i) \varepsilon_0^{1/2}(\mathbf{r}_i) G_0(\mathbf{r}, \mathbf{r}_i) [\mathbf{m}_i \mathbf{n}_i] E_{i-1}(\mathbf{r}_i) d\mathbf{r}_i. \tag{3}$$

Here ε_0 and ε' are the mean and fluctuational parts of the scalar permittivity; $G_0(\mathbf{r}, \mathbf{r}_i)$ is the Green function for the wave equation in the smooth-inhomogeneity approximation[4]; and the vector

$$\mathbf{m}_i = \mathbf{m}_{i-1} - \mathbf{n}_i(\mathbf{m}_{i-1}\mathbf{n}_i) \tag{4}$$

determines the direction of the polarization of the i-tuply scattered wave. The expression (4) is a recursion relation between the polarizations of successive scattering orders.

For the i-fold scattered radiation intensity (1) we obtain with allowance for (2)–(4) the expression

$$I_i(\mathbf{r}) = (k_0^2)^2 \int_V \int_V G_0(\mathbf{r}, \mathbf{r}_i) G_0^*(\mathbf{r}, \mathbf{r}_i') \times \left\{ 1 - \frac{(\mathbf{n}_i \mathbf{m}_{i-1})^2}{|\mathbf{m}_{i-1}|^2} \right\} \langle \varepsilon'(\mathbf{r}_i) \varepsilon'(\mathbf{r}_i') J_{i-1}(\mathbf{r}_i, \mathbf{r}_i') \rangle d\mathbf{r}_i d\mathbf{r}_i',$$
$$J_{i-1}(\mathbf{r}_i, \mathbf{r}_i') = \frac{c}{8\pi} \varepsilon_0^{1/2}(\mathbf{r}_i') \operatorname{Re}(E_{i-1}(\mathbf{r}_i) E_{i-1}^*(\mathbf{r}_i')), \tag{5}$$

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where J_{i-1} is the unaveraged $(i-1)$-fold scattered radiation intensity, which is a functional of ε'.

To carry out the subsequent calculations, we need to uncouple the correlator figuring in (5), and, generally speaking, directly connected with the unknown $2i$-point density-fluctuation correlation function

$$g_{2i}=\left\langle \prod_{l=1}^{i} \Delta\rho(\mathbf{r}_l)\Delta\rho(\mathbf{r}_l') \right\rangle .$$

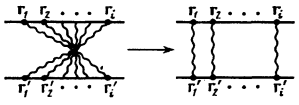
The use of the wave-zone condition in the electrodynamic calculation of the scattered fields (2) and (3) allows us to employ for this purpose the asymptotic formulas obtained in Refs. 20 and 21 for the correlation functions, and corresponding to the elimination of the fluctuation pairs at distances.

$$|\mathbf{r}_l-\mathbf{r}_k|\approx|\mathbf{r}_l'-\mathbf{r}_k'|\gg[k_0\varepsilon_0^{1/2}]^{-1}\approx\lambda. \tag{6}$$

As in the case of double scattering,[4] we can show that the contribution of the non-Gaussian part (of the correlation deviation).

$$g_{2i}-\prod_{l=1}^{i} g_2(\mathbf{r}_l,\mathbf{r}_l')$$

to the intensity of the i-fold scattering is negligibly small, and this virtually allows us to use the previously-obtained[22] Gaussian-type local-equilibrium density-fluctuation distribution function to compute the correlator in (5). In the diagrammatic language[23,5] this approximation for the expression (5) corresponds to the transition from a diagram of the general type with a "correlation star," representing the correlation function g_{2i}, to a diagram of the "ladder" type:



(7)

In (7) a straight line between two neighboring vertices represents the propagator $(\nabla\nabla+k_0^2\hat{I})G_0$; the vertices of the diagram correspond to the density fluctuation; a wavy line corresponds to the correlation interaction of a pair of fluctuations over whose coordinates the integration is performed.

It is clear that the method used to uncouple the correlator in (5) is inapplicable if

$$R_c k_0>1, \tag{8}$$

a condition which is fulfilled when $\tau<(\lambda/2\pi a_0)^{-\nu}\approx 10^{-5}$, where $a_0\approx 10^{-8}$ cm is the correlation length at points far from the critical point.

Taking (7) into account, we can reduce the expression (5) for the intensity of the i-fold scattering in the volume V to the form

$$I_i(\mathbf{r})=I_0\left(\frac{\pi^2}{\lambda^4}\right)^i\frac{V^i}{L^2}\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_\tau^{2i}\int\left\{\prod_{k=1}^{i} g_2(\mathbf{q}_k,\mathbf{r}_k')\left(1-\frac{(\mathbf{n}_k\mathbf{m}_{k-1})^2}{|\mathbf{m}_{k-1}|^2}\right)\right\}\times\prod_{k=1}^{i-1}\frac{d\mathbf{r}_k}{|\mathbf{r}'_{k+1}-\mathbf{r}_k'|^2} \tag{9}$$

$$\mathbf{q}_k=k_0\varepsilon_0^{1/2}(\mathbf{r}_k')(\mathbf{n}_k-\mathbf{n}_{k-1}),$$

where $g_2(\mathbf{q}_k,\mathbf{r}_k')$ is the Fourier transform of the pair correlation function for an inhomogeneous medium[22] with respect to the relative variable $\mathbf{r}_k-\mathbf{r}_k'$; $\mathbf{q}_k$ is the wave vector transferred in the k-th scattering event; and V is the scattering volume.

Let us, by analogy with the single-scattering coefficient,[2] introduce in (9) the coefficient of scattering of arbitrary order:

$$R_i=\frac{I_i}{I_0}\frac{L^2}{V}. \tag{10}$$

Then after performing the integration over the relative distances between the successive scattering points, we obtain the following recursion formula for the scattering coefficients:

$$R_i(\sigma,\sigma_0)=L_0\int R_1(\sigma,\sigma_{i-1})R_{i-1}(\sigma_0,\sigma_{i-1})\,d\sigma_{i-1},\quad i\geqslant 2, \tag{11}$$

$$R_1(\sigma,\sigma_{i-1})=\frac{V\pi^2}{\lambda^4}\left(\rho\frac{\partial\varepsilon}{\partial\rho}\right)_\tau^2 g_2(\mathbf{q}_i)\left[1-\frac{(\mathbf{n}_i\mathbf{m}_{i-1})^2}{|\mathbf{m}_{i-1}|^2}\right]. \tag{12}$$

The successive integration in (11) over the solid angles σ_k sums the scattered radiation of all multiplicities $k<i$ (which is the source of the i-fold scattered radiation, and is detected by the receiver) in the interior of the medium over the directions.

The quantity L_0, which has the dimensionality of length (in the general case a function of the linear dimensions of the system), depends on the geometry of the problem, and is an important parameter of the "instrumental" theory of multiple scattering, which is adequate for a specific experiment (see, for example, Refs. 6 and 15). In particular, for the investigation of scattering in a volume having the shape of a sphere of radius R', we have $L_0=R'$, whereas for a cube with edge a uniformly illuminated through one of its faces, the quantity L_0 is equal to $a\sqrt{3}/2$.

In view of the increased interest in the experimental investigations[6,9,10] of the dependence of double (multiple) scattering on the characteristic linear dimensions of the scattering volume, let us note that, on the basis of the relations (10) and (11), $R_i\sim L_0^{i-1}$. Hence for the intensity of the i-fold scattering we have $I_i\sim VL_0^{i-1}$, which has been noted before.[12] The result $I_2\sim VL_0$ was theoretically obtained[3-5] in the solution of the electrodynamic problem of multiple scattering, and the result $I_i/I_{i-1}\sim L_0$ $(i\geqslant 2)$ was obtained in Ref. 24 in an investigation of multiple scattering by the radiation-transport-equation method.

2. Let us consider the total multiple-scattering coefficient

$$R=\sum_{i=1}^{\infty} R_i. \tag{13}$$

Taking (9)–(12) into account, we easily find that the expression (13) is an iterative solution to the integral equation

$$R(\sigma,\sigma_0)=R_1(\sigma,\sigma_0)+L_0\int R_1(\sigma,\sigma')R(\sigma',\sigma_0)\,d\sigma'. \tag{14}$$

The analysis of the total multiple-scattering coefficient R in the form of the iterative series (13) meets with considerable difficulties. Of immediate interest is the obtaining of a closed solution to Eq. (14). Let us rep-

resent the sought solution in the form of a series in terms of the complete system of spherical functions:

$$R(\sigma,\sigma_0)=\sum_{n=0}^{\infty}\sum_{m=-n}^{n}A_{mn}Y_n^m(\sigma),\qquad Y_n^m(\sigma)=\left[\left(\frac{2n+1}{4\pi}\right)\frac{(n-m)!}{(n+m)!}\right]^{1/2}P_n^m(\cos\vartheta)e^{im\varphi}, \tag{15}$$

where the $P_n^m(\cos\vartheta)$ are the associated Legendre polynomials. Correspondingly,

$$R_1(\sigma,\sigma_0)=\sum_{m,n}B_{mn}Y_n^m(\sigma), \tag{16a}$$

$$R_1(\sigma,\sigma')=\sum_{m,n}\sum_{m',n'}C_{mn,m'n'}Y_n^m(\sigma)Y_{n'}^{m'*}(\sigma'). \tag{16b}$$

Let us note that, because the scattering coefficients R_1 and R are red, the coefficients

$$A_{mn}=\int R(\sigma,\sigma_0)Y_n^{m*}(\sigma)\,d\sigma, \tag{17a}$$

$$B_{mn}=\int R_1(\sigma,\sigma_0)Y_n^{m*}(\sigma)\,d\sigma, \tag{17b}$$

$$C_{mn,m'n'}=\iint R_1(\sigma,\sigma')Y_n^{m*}(\sigma)Y_{n'}^{m'}(\sigma')\,d\sigma\,d\sigma' \tag{17c}$$

of the expansions (15) and (16) satisfy the relations

$$A_{mn}=A_{-mn}^{*},\quad B_{mn}=B_{-mn}^{*},\quad C_{mn,m'n'}=C_{-mn,-m'n'}^{*}. \tag{18}$$

Using (14)–(16) and the orthogonality condition for the spherical functions

$$\int Y_n^m(\sigma)Y_{n'}^{m'*}(\sigma)\,d\sigma=\delta_{mm'}\delta_{nn'}, \tag{19}$$

we obtain for the determination of the expansion coefficients A_{mn} the following system of algebraic equations:

$$A_{mn}=B_{mn}+L_0^{*}\sum_{m',n'}C_{mn,m'n'}A_{m'n'}. \tag{20}$$

It follows from (17b) and (17c) that the form of the coefficients B_{mn} and $C_{mn,m'n'}$, and, consequently, of the A_{mn}, depends essentially on the structure of the Fourier transform of the correlation function g_2 [see (12)]. At the same time, using (12) and (17b), we can show that only the coefficients B_{0n} and B_{2n} are nonzero. This selection rule for the coefficients of the expansion (16a) follows directly after the integration over the azimuthal direction in (17b), and is not connected with the specific structure of the pair correlation function. Taking account of (4), (12), and (19), we obtain for the coefficients $C_{mn,m'n'}$ from (17c) in an entirely similar fashion the relation

$$C_{mn,m'n'}=B_{mn}'\delta_{m0}\delta_{m'm}\delta_{n'n},\qquad B_{mn}'=\left(\frac{4\pi}{2n+1}\right)^{1/2}B_{mn}.$$

Taking the above-noted properties of the coefficients B_{mn} and $C_{mn,m'n'}$ into account, we easily obtain from (20) the solution for the coefficients A_{mn}:

$$A_{mn}=B_{mn}(\delta_{m0}+\delta_{m2})/(1-L_0B_{0n}'),\quad n\geqslant m. \tag{21}$$

Then, using the condition (18), we can rewrite the expansion (15) for the scattering coefficient R in the form

$$R(\sigma,\sigma_0)=\sum_{n\geqslant 0}\{[2B_{0n}Y_n^0(\sigma)+B_{2n}(Y_n^2(\sigma)+Y_n^{-2}(\sigma))]/2(1-L_0B_{0n}')\}. \tag{22}$$

The formula (22) is convenient in that it enables us to find the various characteristics of multiple scattering (the indicatrix, the extinction coefficient, their temperature and field characteristics, the correlation properties of the dispersive medium) from the given correlation function g_2.

Let us note that, according to (17b), the coefficient B_{00} is the extinction coefficient k_1 of the single-scattering approximation ($k_1=B_{00}$). From the formula (22) we directly obtain the criterion for the applicability of the iterative procedure used above in the derivation of Eq. (14), namely,

$$k_1L_0<1 \tag{23}$$

(for specific numerical estimates for this criterion, see Sec. 3).

The extinction coefficient K for multiple scattering is given by the expression

$$K=A_{00}=\int R(\sigma,\sigma_0)\,d\sigma, \tag{24}$$

which, after the use of (19) and (22), turns out to be equal to

$$K=k_1/(1-k_1L_0). \tag{25}$$

The expression (25) can also be obtained from the series (13). Indeed, using the recursion formula (11), and performing the integration over the solid angle, we find in accordance with the definition (24) that

$$K=k_1+k_1^2L_0+k_1^3L_0^2+\ldots. \tag{26}$$

The summation of the series (26) with allowance for the criterion (23) leads to the formula (25).

From (26) we obtain the following important relation between the contributions of the successive scattering orders to the multiple-scattering extinction coefficient:

$$k_{i+1}/k_i=k_1L_0\quad(i=1,2,\ldots), \tag{27}$$

which is given in our last paper,[19] where we discuss, in particular, the thermodynamic conditions under which the corrections for multiple scattering are important.

When the strong inequality $k_1L_0\ll 1$ is fulfilled, the experimentally determinable quantity is, as follows from (25), the single-scattering extinction coefficient k_1. But in the general case what is determined in experiment is not at all the quantity k_1, but the multiple-scattering extinction coefficient K. Solving (25) for k_1,

$$k_1=K/(1+KL_0), \tag{28}$$

and substituting into the relation (27), we obtain

$$k_{i+1}/k_i=KL_0/(1+KL_0)\quad(i=1,2,\ldots). \tag{29}$$

The formula (29) enables us to find the relative contributions of the successive scattering orders in terms of the experimentally determinable quantity K, the extinction coefficient.

Let us briefly discuss the possibility of an experimental determination of the extinction coefficient K. The traditional approach is based on the application of the Bouguer–Lambert law

$$I(l)=I_0\exp(-k'l) \tag{30}$$

for the intensity $I(l)$ of a light beam that has traversed

a distance l in the medium and the identification of the reciprocal attenuation distance k' with extinction coefficient. It is natural that, in the case in which fundamental absorption of the electromagnetic waves does not occur in the medium, the decrease of the intensity is entirely due to the scattering.

The basis for the use of the expression (30) is the well-known optical theorem (see, for example, Ref. 25), the region of applicability of which is limited by the single-scattering approximation ($k' = k_1$). If this approximation ceases to be applicable, then it becomes necessary to relate the reciprocal attenuation distance k' already introduced formally in (30) with the multiple-scattering extinction coefficient K.

For a light-diffusing medium, we have, when the surface effects are neglected, the following balance equation:

$$\sigma^* I_0 = \sigma^* I(l) + I_0 V K, \tag{31}$$

where σ^* is the cross-section area of the incident (transmitted) light beam. Approximating the intensity of the transmitted beam by the formula (30), we obtain the sought relation between the reciprocal attenuation distance k' and the multiple-scattering extinction coefficient K:

$$k' = l^{-1} \ln (1 - VK/\sigma^*). \tag{32}$$

Let us note that it does not at all follow from the smallness of the experimentally determinable—with the aid of the formula (30)—quantity $k'l$ that k' is the single-scattering extinction coefficient k_1. Indeed, in the case $k'l \ll 1$ we find from (32) that

$$K = \sigma^* l k'/V. \tag{33}$$

Hence when the condition $\sigma^* l/V = 1$ is fulfilled, the experimentally determinable reciprocal attenuation distance k' coincides with the multiple-scattering extinction coefficient K. This result can be regarded as a generalization of the optical theorem to the case of multiple scattering. In the general case, in which $\sigma^* l/V \neq 1$, the simultaneous use of the formulas (29) and (33) allows us to estimate the ratio of the intensities of successive scattering orders from the experimentally measurable quantity k' and the known geometry factors l, σ^*, V, and L_0.

In the case in which the extinction coefficient K found from (33) satisfies the condition $KL_0 \ll 1$, we find on the basis of (28) that $k_1 = K$, i.e., that the dominant contribution to k' is made by single scattering (in the single-scattering approximation the condition $\sigma^* l = V$ is fulfilled automatically). It is in this, and only in this, case that the optical theorem is valid and the reciprocal attenuation distance k' in the Bouguer–Lambert law (30) is the single-scattering extinction coefficient k_1.

For the elucidation of the presence of the contributions of the higher-scattering orders in terms of the experimentally measured value of the reciprocal attenuation distance k' and the known geometry factors l, σ^*, V, and L_0, we can use the following method. We should, by varying the distance l traversed by the light beam in the medium, determine the linear segment in the plot of $\ln[I_0/I(l)]$ against l. The slope of this graph gives the single-scattering extinction coefficient k_1. The deviation from linearity indicates the presence of higher scattering orders. The reciprocal attenuation distance determined on the basis of (30) and (31) becomes a function of not only the thermodynamic variables, but also the geometry factors, which indicates that k' cannot be identified with the single-scattering extinction coefficient k_1.

3. For the performance of specific calculations of the multiple-scattering extinction coefficient K, we shall use below the correlation function $g_2(\mathbf{q}, \mathbf{r}')$ of the Ornstein–Zernike approximation for a spatially inhomogeneous fluid near the critical point in a gravitational field[22]:

$$g_2(\mathbf{q}, z) = \frac{k_B T}{V b p_c} \frac{1}{R_c^{-2}(z, \tau) + q^2}. \tag{34}$$

In this case for the single-scattering coefficient (12) we have

$$R_1(\sigma, \sigma_0) = 2\pi\alpha(z, \tau) \frac{1 - (\mathbf{n}_1 \mathbf{m}_0)^2}{1 + \delta(z, \tau)(1 - \mathbf{n}_1 \mathbf{n}_0)},$$
$$\alpha(z, \tau) \sim \beta_T(z, \tau)/\lambda^4, \quad \delta(z, \tau) = q^2 R_c^2(z, \tau)\ [4], \quad q = 2(2\varepsilon)^{1/2}\pi/\lambda, \tag{35}$$

where q is the wave vector transferred in a scattering through an angle $\vartheta = \pi/2$.

The expansion coefficients B_{0n} and B_{2n} entering into the expression (22) are given by the following formulas:

(36)

$$B_{0n} = 4(2n+1)^{1/2}\pi^2 \frac{\alpha}{\delta} \left\{ \left[\left(\frac{1+\delta}{\delta} \right)^2 + 1 \right] Q_n\left(\frac{1+\delta}{\delta} \right) - \frac{1+\delta}{\delta} \delta_{n0} - \frac{1}{3} \delta_{n1} \right\},$$
$$B_{2n} = 2 \left[(2n+1) \frac{(n-2)!}{(n+2)!} \right]^{1/2} \pi^2 \frac{\alpha}{\delta} \left\{ 2 \frac{1+\delta}{\delta} Q_{n+1}\left(\frac{1+\delta}{\delta} \right) - \left[\left(\frac{1+\delta}{\delta} \right)^2 (n+2) - n \right] Q_n\left(\frac{1+\delta}{\delta} \right) \right\}, \tag{37}$$
$$n \geqslant 2,$$

where the $Q_n(x)$ are generalized Legendre polynomials.

From (36) we obtain for the single-scattering extinction coefficient the well-known expression (see also Refs. 26–28)

$$k_1 = B_{00} = 2\pi^2 \frac{\alpha}{\delta} \left\{ \frac{1 + 2\delta + 2\delta^2}{\delta^2} \ln(1 + 2\delta) - \frac{2(1+\delta)}{\delta} \right\}. \tag{38}$$

It follows from the formula (38) that, as the critical point is approached, the extinction coefficient slowly increases, and diverges logarithmically as $\delta \to \infty$. This circumstance leads, generally speaking, to the violation of the criterion (23) for the convergence of the iterative procedure.

Let us determine those values of the thermodynamic variables (or the parameter δ) and the geometry factor L_0 for which the criterion (23) is still fulfilled. For this purpose we carried out a numerical computation of the quantity

$$\frac{k_1 L_0}{s} = \pi \left\{ \frac{1 + 2\delta + 2\delta^2}{\delta^2} \ln(1 + 2\delta) - \frac{2(1+\delta)}{\delta} \right\},$$
$$s = \left(\rho \frac{\partial \varepsilon}{\partial \rho} \right)_T^2 k_B T L_0 / 8\lambda^2 f', \tag{39}$$

where s is a factor that was introduced earlier[19] in the

computation of the depolarization coefficient Δ and f^* is a parameter characterizing the nonlocalized nature of the fluctuations.[2] The calculation was performed in the δ-parameter range from 10^{-3} to 1.0 in steps of 10^{-3} and in the range $1.0 \leq \delta \leq 11.0$ in steps of 10^{-2}. Some of the computational data are given in Table I.

For the realistic parameter $s = 0.1$ ($L_0 \approx 10^{-2}$ cm for Xe) the condition (23) is violated ($k_1 L_0 = 1$) when $\delta = 4.15$, which corresponds to $qR_c \approx 2$ and a temperature deviation $\tau \approx 10^{-4.9}$ along the critical isochore. For the same value of s the single-scattering approximation can be used for $\tau \gtrsim 10^{-3.7}$ with acceptable accuracy when the ratio k_{i+1}/k_i of the contributions of successive scattering orders to the extinction coefficient does not exceed 10%. These results agree with the conclusion, drawn in a number of experimental investigations,[14,15] that the higher-order scatterings in the critical region become important when $qR_c \gtrsim 1$.

The above-performed computation allows us to determine more accurately the limits of applicability of the results of the computations, carried out in the double-scattering approximation, of the depolarization coefficient Δ. As has been shown,[19] for $s = 0.1$, the coefficient Δ assumes its maximum value when $\delta_{max} = 7.8$ ($\tau \approx 10^{-5.16}$), and then begins to decrease, which indicates the inexpediency of a further depolarization-coefficient calculation in the double-scattering approximation and an increase in the role of the higher-scattering orders when $\delta > \delta_{max}$.

The possibility of the summation of all the scattering orders corresponding to the ladder diagrams of the type (7) furnishes a more exact limit of applicability of the scattering-order concept. In the model calculation based on the use of the Ornstein–Zernike approximation, this limit is characterized by the fulfillment of the inequality

$$\frac{\pi(\rho\partial\varepsilon/\partial\rho)_T^2 k_B T L_0}{8\pi^2 f^*}\left\{\frac{1+2\delta+2\delta^2}{\delta^2}\ln(1+2\delta)-\frac{2(1+\delta)}{\delta}\right\}<1. \qquad (40)$$

The inequality (40) can be secured by an appropriate choice of the geometry factor L_0 or of the thermodynamic variables τ and $\Delta\rho = (\rho - \rho_c)/\rho_c$ (τ and the dimensionless height z in an experiment with allowance for the gravitational effect).

4. In conclusion, let us note that the iterative procedure used in the solution of the electrodynamic problem corresponds to the well-known Born expansion in the quantum-mechanical theory of scattering (see, for example, Ref. 29). The order concept for the scattered field is then determined by the number of the functional factors $\varepsilon'(\mathbf{r}_i) \sim \Delta\rho(\mathbf{r}_i)$ in each term of the iteration series, while the order of the scattered-light intensity is (when the non-Gaussian contributions are neglected) determined by the number of functional factors $g_2(\mathbf{r}_i, \mathbf{r}_i')$ that correspond to the correlation interaction of the density fluctuations at the symmetrical vertices on the lines of the diagrams. The summation of the series in the order for the intensity becomes possible as a result of the use of the wave-zone condition, which allows us to select the periodic solutions of the wave equation for each of the scattering centers. A class of ladder diagrams in which the distances between neighboring vertices on each line staisfy the condition (6) then separates out from the entire diagram series. The investigation, performed in this approximation, of the extinction coefficient K shows that the inequality (23) is violated as the critical point is approached. The scattering-order concept itself thus loses its meaning.

Further refinement of the description of the process of light scattering in the critical region requires a more consistent approach based on the technique of resummation of the basic series for the fields and the intensity of the scattered radiation, as well as the use of functional methods to solve the electromagnetic problem in the case of non-Gaussian order-parameter-fluctuation statistics.

TABLE I.

δ	10^{-3}	10^{-2}	10^{-1}	$2\cdot10^{-1}$	$3\cdot10^{-1}$	$4\cdot10^{-1}$	$5\cdot10^{-1}$	$6\cdot10^{-1}$	$7\cdot10^{-1}$	$8\cdot10^{-1}$	$9\cdot10^{-1}$
$-\lg\tau$	2,001	2.801	3.601	3.842	3.982	4.082	4.160	4.223	4.277	4 323	4.364
$\frac{k_1 L_0}{s}$	$8.372\cdot10^{-3}$	$8.295\cdot10^{-2}$	$7.641\cdot10^{-1}$	1.412	1.976	2.476	2.926	3.336	3.713	4.061	4.386
δ	1,0	2.0	3.0	4,0	5,0	6.0	7.0	8.0	9,0	10.0	11.0
$-\lg\tau$	4 401	4.642	4.782	4.882	4.960	5.023	5.077	5.123	5,164	5.201	5.234
$\frac{k_1 L_0}{s}$	4,691	7.008	8.604	9.834	10.841	11.700	12.439	13.689	13,689	14.226	14.719

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