$$\frac{g_I}{g_I'} = \frac{(A/A')_1}{(\alpha_s/A)_1\Delta_s + 1} = \frac{(A/A')_2}{(\alpha_s/A)_2\Delta_s + 1} ,$$

$$\Delta_s = \frac{(A/A')_1 - (A/A')_2}{(\alpha_s/A)_1(A/A')_2 - (\alpha_s/A)_2(A/A')_1} ,$$

where 1 and 2 refer to different levels. α_s and α are obtained from the relations

$$A(^4F_{9/2}) = \tfrac{1}{9}a_s + \tfrac{1}{3}a_p' + \tfrac{5}{9}a_d' ,$$

$$A(^4P_{5/2}) = 0.199a_s - 0.473a_p' + 0.840a_d' .$$

From the results in Table I we find

$$^{63}\Delta_s{}^{65} = 0.000\,048(5), \quad \frac{g_I{}^{63}}{g_I{}^{65}} = 0.933\,524(5).$$

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Correlation Properties of Light Scattered From Fluids*

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The fluctuation and correlation properties of light scattered by a fluid are studied, without neglect of the spectral linewidth of the incident field. When the source is a single-mode laser, it is shown that, although the instantaneous amplitude of the scattered field obeys a Gaussian probability density, the scattered field is not a Gaussian field. The linewidth of the laser beam is reflected in the amplitude correlations of the scattered light, but not in the intensity correlations. On the other hand, when the laser is oscillating in more than one mode simultaneously, the spectral profile of the laser beam makes a contribution to the spectral density of the scattered intensity fluctuations, and cannot be neglected.

1. INTRODUCTION

The problem of light scattering by a fluid, particularly near the critical point, has been the subject of a great many experimental[1–14] and theoretical[14–25,4,8] investigations since the early work of Brillouin.[26] In recent years additional interest in the field was stimulated by the light beating technique developed by Benedek[3,4,6,7] and his coworkers, although light beating experiments were first reported by Forrester, Gudmundsen, and Johnson[27] in 1955. It has been demonstrated that measurements of the frequency shifts and the scattering angles are capable of yielding valuable information about the correlation properties of the fluid scatterers. But, while the fluid correlations have been treated in considerable detail, the coherence properties of the incident light have tended to be disregarded almost completely. It has usually been assumed that the incident field is completely monochromatic, or completely coherent, or both, so that the correlations of the scattered field reflect the properties of the scatterers and nothing else. As a consequence, it is frequently taken for granted that the scattered field may be treated as a Gaussian random process.

While it is true that the light beam from a single-mode laser can be almost completely spatially coherent and has a long coherence time, the coherence time is always finite. Indeed, the frequency spread of the incident light beam is usually much greater than the frequency shifts introduced by the scattering process. The coherence properties of the incident field can therefore not be disregarded, even if it is a laser field.

In the following we examine the correlation properties of the scattered field on the basis of some reasonable assumptions about the fluctuations of the laser field and of the fluid scatterers. No theoretical treatment of the fluid correlations is attempted. We show explicitly that, while the probability density of the instantaneous scattered wave amplitude is Gaussian, the scattered field is *not* a Gaussian random process, as has often been assumed. The amplitude and intensity correlation functions of the scattered field in general do not satisfy the relationship expected for a Gaussian field. The linewidth of the incident laser beam is strongly reflected in the amplitude correlation of the scattered light, but not in the intensity correlation, if a single-mode laser is used. We find, however, that the formula which is usually derived on the basis of the Gaussian assumption, with the further assumption of zero linewidth for the laser beam, is correct, since the effects of these assumptions cancel.

On the other hand, if the laser is oscillating in two or more modes simultaneously, the spectral profile of the laser beam makes an explicit contribution to the intensity correlation of the scattered light. Particularly if the laser beam contains a number of off-axis modes, the laser's spectral profile may distort the measured spectral density. This is illustrated by an example in Sec. 6.

2. REPRESENTATION OF THE SCATTERED FIELD

We suppose that the incident field is in the form of a plane beam falling on a medium whose susceptibility fluctuates both in space and time. The incident beam induces an oscillating polarization which causes the medium to radiate, and we refer to the radiation from the susceptibility fluctuations as the scattered field. Since quantum properties of the field do not play any significant role in the problem, we treat the radiation field classically throughout.

It is convenient to make a plane wave expansion of the incident field $\vec{E}_0(\vec{x}, t)$ in the usual form

$$\vec{E}_0(\vec{x}, t) = \sum_s \int d^3k\, \vec{\epsilon}_{\vec{k}, s}[v_{\vec{k}, s} \exp i(\vec{k}\cdot\vec{x} - ckt) + \text{c. c.}], \tag{1}$$

where the wave vector $\vec{k}$ and polarization index s label the modes of the field, and $\vec{\epsilon}_{\vec{k}, s}$ is the unit polarization vector. The field is assumed to be nearly plane and quasimonochromatic, so that $v_{\vec{k}, s}$ vanishes with high probability except for wave vectors pointing in the same direction, whose magnitudes lie within a small range Δk centered on k_0. If the susceptibility $\chi(\vec{x}, t)$ is a scalar, then the induced polarization at each element d^3x of scatterer is

$$\chi(\vec{x}, t)\vec{E}_0(\vec{x}, t)d^3x \quad ,$$

and the scattered field $\vec{E}(\vec{x}, t)$ radiated by this polarization at some distant point $\vec{X}$, is given by the usual dipole formula[28] for each Fourier component of Eq. (1),

$$\vec{E}(\vec{X}, t) = -\int_{\mathcal{V}} d^3x R^{-1} \Delta\chi(\vec{x}, t - R/c) \sum_s \int d^3k\, k^2 [v_{\vec{k}, s} \exp i(\vec{k}\cdot\vec{x} - ckt) + \text{c. c.}]\, \vec{\epsilon}_{\vec{k}, s} \cdot (\vec{\rho}\vec{\rho} - 1) \quad . \tag{2}$$

Here $\vec{\rho}$ is the unit vector in the direction $\vec{X} - \vec{x}$, $R = |\vec{X} - \vec{x}|$, and the space integral is to be taken over the volume $\mathcal{V}$ of the scatterer. $\Delta\chi(\vec{x}, t)$ is the deviation of $\chi(\vec{x}, t)$ from its mean value $\langle\chi(\vec{x}, t)\rangle$, which we take to be independent of space and time within the scattering region $\mathcal{V}$.

In addition to the scattered field given by Eq. (2), the incident, or diffracted, field may also make a contribution at $(\vec{X}, t)$, but this contribution can be made small by suitable choice of $\vec{X}$, and can also be distinguished experimentally from the scattered field, and we shall not be concerned with it here. A simplification which may appear to be completely unjustified is the neglect of any difference of refractive index between the scattering region and the surrounding space in Eq. (2). A moment's thought will show that the effect of a significant index difference is to introduce very complicated geometric corrections to the phase of $\vec{E}(\vec{X}, t)$, but that the degree of amplitude correlation, and any intensity correlation, will not be affected.

In view of the quasimonochromaticity of the incident field, the factor k^2 may be treated as nearly constant under the integral in Eq. (2) and replaced by k_0^2. Finally, if the incident field is linearly polarized at right angles to the scattering plane, i.e., normal to both $\vec{k}$ and $\vec{\rho}$, then $\vec{\epsilon}_{\vec{k}, s}\cdot\vec{\rho}=0$, and Eq. (2) reduces to

$$\vec{E}(\vec{X}, t)=k_0^2\int_{\mathcal{V}} d^3x R^{-1}\Delta\chi(\vec{x}, t-R/c)\vec{E}_0(\vec{x}, t-R/c) \quad . \tag{3}$$

We now introduce the analytic signal representation of the fields,[29] by making a one-dimensional Fourier expansion of both $\Delta\chi(\vec{x}, t)$ and $\vec{E}_0(\vec{x}, t)$ in the form

$$\Delta\chi(\vec{x}, t)=\int_{-\infty}^{\infty}\Delta\psi(\vec{x}, \omega)e^{-i\omega t}d\omega \quad , \quad \vec{E}_0(\vec{x}, t)=\int_{-\infty}^{\infty}\vec{u}(\vec{x}, \omega)e^{-i\omega t}d\omega \quad , \tag{4}$$

so that

$$\vec{E}(\vec{X}, t)=k_0^2\int_{\mathcal{V}} d^3x R^{-1}\iint_{-\infty}^{\infty} d\omega' d\omega\, \Delta\psi(\vec{x}, \omega')\vec{u}(\vec{x}, \omega)\exp[-i(\omega+\omega')(t-R/c)]$$

$$=k_0^2\int_{\mathcal{V}} d^3x R^{-1}\iint_{-\infty}^{\infty} d\omega'' d\omega\, \Delta\psi(\vec{x}, \omega''-\omega)\vec{u}(\vec{x}, \omega)\exp[-i\omega''(t-R/c)] \quad , \tag{5}$$

when we make the substitution $\omega+\omega'=\omega''$. The analytic signal representation $\vec{V}(\vec{X},t)$ then follows if we suppress the negative frequency range of ω'', so that

$$\vec{V}(\vec{X}, t)=k_0^2\int_{\mathcal{V}} d^3x R^{-1}\int_0^{\infty} d\omega''\int_{-\infty}^{\infty} d\omega\, \Delta\psi(\vec{x}, \omega''-\omega)\vec{u}(\vec{x}, \omega)\exp[-i\omega''(t-R/c)] \,. \tag{6}$$

We now observe that, while $\vec{u}(\vec{x}, \omega)$ is appreciably different from zero only for high (optical) frequencies $|\omega|\sim k_0$, $\Delta\psi(\vec{x}, \omega)$ is appreciably different from zero only for low frequencies of some thousands cps,[3–5,7–9,13,14] since the susceptibility fluctuates slowly. It follows that the negative range of the ω integral makes a negligible contribution and can be disregarded. The inverse substitution $\omega''-\omega=\omega'$ then leads to

$$\vec{V}(\vec{X}, t)=k_0^2\int_{\mathcal{V}} d^3x R^{-1}\int_0^{\infty} d\omega\int_{-\omega}^{\infty} d\omega'\Delta\psi(\vec{x}, \omega')\vec{u}(\vec{x}, \omega)\exp[-i(\omega+\omega')(t-R/c)]$$

$$=k_0^2\int_{\mathcal{V}} d^3x R^{-1}\Delta\chi(\vec{x}, t-R/c)\vec{V}_0(\vec{x}, t-R/c) \quad , \tag{7}$$

where $\vec{V}_0(\vec{x}, t)$ is the analytic signal corresponding to $\vec{E}_0(\vec{x}, t)$.

Finally we observe that, since the amplitude of a single-mode laser beam (far above threshold) may be treated as constant to a very good approximation, we can substitute

$$\vec{V}_0(\vec{x}, t)=\vec{\epsilon}(I_0)^{1/2}\exp i[\vec{k}_0\cdot\vec{x}-ck_0t-\beta(\vec{x}, t)] \tag{8}$$

in Eq. (7). Here the phase $\beta(\vec{x}, t)$ is a slowly varying, random function of space and time, $I_0=\vec{V}_0^*(\vec{x}, t)\cdot\vec{V}_0(\vec{x}, t)$ is the mean light intensity, $\vec{k}_0$ is the wave vector of magnitude k_0 in the direction of propagation, and it is assumed that the linear dimensions of $\mathcal{V}$ are small compared with the coherence length $c/\Delta k$ of the incident light. We can therefore treat $\beta(\vec{x}, t)$ as constant under the integral in Eq. (7), and obtain

$$\vec{V}(\vec{X}, t)=\vec{\epsilon}(I_0)^{1/2}k_0^2\exp[-i\beta(\vec{x}_0, t-R_0/c)]\int_{\mathcal{V}} d^3x R^{-1}\Delta\chi(\vec{x}, t-R/c)\exp i[\vec{k}_0\cdot\vec{x}-ck_0(t-R/c)] \quad , \tag{9}$$

where $\vec{x}_0$ is the midpoint of $\mathcal{V}$ and $R_0=|\vec{X}-\vec{x}_0|$.

3. PROBABILITY DENSITY OF THE SCATTERED AMPLITUDE

In order to establish the probability density of the scattered complex wave amplitude $\vec{V}(\vec{X}, t)$, we make the assumption that, since the susceptibility fluctuations are produced by the random motions of particles, $\Delta\chi$ in Eq. (9) may be treated as a Gaussian random process.[30] This implies that the joint probability density for $\Delta\chi$, evaluated at any number of distinct space-time points, may be taken to be a multivariate Gaussian function. This conclusion is implicit in many of the theoretical treatments of the scattering problem. β is a random phase angle which we take to be distributed uniformly over the range 0 to 2π. We may therefore express the real part $\vec{E}(\vec{X}, t)$ of $\vec{V}(\vec{X},t)$ in the form

$$\vec{E}=\vec{\epsilon}Q\cos(\alpha-\beta) \quad , \tag{10}$$

where

$$Q=(A^2+B^2)^{1/2}, \quad \tan\alpha = B/A \ ,$$
$$A=(I_0)^{1/2}k_0^2\int_{\mathcal{V}} d^3x\,R^{-1}\Delta\chi(\vec{x}, t-R/c)\cos[\vec{k}_0\cdot\vec{x}-ck_0(t-R/c)] \ , \tag{11}$$
$$B=(I_0)^{1/2}k_0^2\int_{\mathcal{V}} d^3x\,R^{-1}\Delta\chi(\vec{x}, t-R/c)\sin[\vec{k}_0\cdot\vec{x}-ck_0(t-R/c)] \ ,$$

and A and B are themselves Gaussian random processes of zero mean, by virtue of the Gaussian nature of $\Delta\chi$.

In order to show that α is almost uniformly distributed over the range 0 to 2π, we note that A and B have the same variance but are practically uncorrelated. For, if the factor R^{-1} may be treated as nearly constant under the integral, we have

$$\begin{aligned}\langle A^2\rangle &= (I_0k_0^4/R_0^2)\int\int_{\mathcal{V}} d^3x\,d^3x'\langle\Delta\chi(\vec{x}, t-R/c)\Delta\chi(\vec{x}', t-R'/c)\rangle \\ &\quad\times\cos[\vec{k}_0\cdot\vec{x}-ck_0(t-R/c)]\cos[\vec{k}_0\cdot\vec{x}'-ck_0(t-R'/c)] \\ &= (I_0k_0^4/R_0^2)\int_{\mathcal{V}} d^3x\int d^3x''\langle\Delta\chi(\vec{x}, t-R/c)\Delta\chi(\vec{x}+\vec{x}'', t-R'/c)\rangle \\ &\quad\times\cos[\vec{k}_0\cdot\vec{x}-ck_0(t-R/c)]\cos[\vec{k}_0\cdot(\vec{x}+\vec{x}'')-ck_0(t-R'/c)] \ ,\end{aligned} \tag{12}$$

when we substitute $\vec{x}'-\vec{x}=\vec{x}''$, provided the correlation range of $\Delta\chi$ is very small compared with the linear dimensions of $\mathcal{V}$. It seems reasonable to suppose that the susceptibility fluctuations are not only stationary in time, but also homogeneous over the scattering volume $\mathcal{V}$, so that we may write

$$\langle\Delta\chi(\vec{x}, t-R/c)\Delta\chi(\vec{x}+\vec{x}'', t-R'/c)\rangle = \mu(\vec{x}'', (R-R')/c) \ . \tag{13a}$$

It is known that $\mu(\vec{x}, \tau)$ is a slowly varying function of τ from measurements[3–5,7–9,13,14] which show that the corresponding spectral density is usually limited to some thousands of cps. Then, for all laboratory-size scatterers, we may put

$$\mu(\vec{x}'', (R-R')/c) = \mu(\vec{x}'', 0). \tag{13b}$$

We now make the far-field approximation, which allows us to express R in the form

$$R=|R_0\vec{\rho}-(\vec{x}-\vec{x}_0)|\approx R_0-\vec{\rho}\cdot(\vec{x}-\vec{x}_0) \ , \tag{14}$$

for sufficiently distant R, where $\vec{x}_0$ is the mid-point of the scattering region, $R_0=|\vec{X}-\vec{x}_0|$, and $\vec{\rho}$ is the unit vector in the direction $\vec{X}-\vec{x}_0$. With the help of Eqs. (13b) and (14) we can now perform the integration over $\vec{x}$ in Eq. (12). For a rectangular volume of linear dimensions l_1, l_2, l_3 we obtain

$$\begin{aligned}\langle A^2\rangle &= \tfrac{1}{2}(I_0k_0^4\mathcal{V}/R_0^2)\int d^3x''\mu(\vec{x}'', 0)\{\cos[(\vec{k}_0+\vec{K})\cdot\vec{x}''] \\ &\quad+\cos[2\vec{k}_0\cdot\vec{x}_0+(\vec{k}_0+\vec{K})\cdot\vec{x}''-2ck_0(t-R_0/c)]\prod_{j=1,2,3}\sin[(\vec{k}_0+\vec{K})_j l_j]/(\vec{k}_0+\vec{K})_j l_j\} \ ,\end{aligned} \tag{15}$$

where $\vec{K}=k_0\vec{\rho}$ is the wave vector of the scattered light received at $\vec{X}$. If l_1, l_2, and l_3 are much greater than typical wavelengths of the light, as we may assume, then the second term is negligible compared with the first, and $\langle A^2\rangle$ is very nearly proportional to the cosine transform of $\mu(\vec{x}'', 0)$.

Similarly, we may show that

$$\begin{aligned}\langle B^2\rangle &= \tfrac{1}{2}(I_0k_0^4\mathcal{V}/R_0^2)\int d^3x''\mu(\vec{x}'', 0)\{\cos[(\vec{k}_0+\vec{K})\cdot\vec{x}''] \\ &\quad-\cos[2\vec{k}_0\cdot\vec{x}_0+(\vec{k}_0+\vec{K})\cdot\vec{x}''-2ck_0(t-R_0/c)]\prod_{j=1,2,3}\sin[(\vec{k}_0+\vec{K})_j l_j]/(\vec{k}_0+\vec{K})_j l_j\} \ ,\end{aligned} \tag{16}$$

so that, to a very good approximation,

$$\langle A^2\rangle = \langle B^2\rangle = K \ . \tag{17}$$

However, for the correlation $\langle AB\rangle$ we find, by a similar argument,

$$\begin{aligned}\langle AB\rangle &= \tfrac{1}{2}(I_0k_0^4\mathcal{V}/R_0^2)\int d^3x''\mu(\vec{x}'', 0)\{\sin[(\vec{k}_0+\vec{K})\cdot\vec{x}''] \\ &\quad+\cos[2\vec{k}_0\cdot\vec{x}_0+(\vec{k}_0+\vec{K})\cdot\vec{x}''-2ck_0(t-R_0/c)]\prod_{j=1,2,3}\sin[(\vec{k}_0+\vec{K})_j l_j]/(\vec{k}_0+\vec{K})_j l_j\} \ .\end{aligned} \tag{18}$$

The contribution of the first term vanishes by virtue of the symmetry of $\mu(\vec{x}'', 0)$, while that of the second term is very small compared with $\langle A^2\rangle$ or $\langle B^2\rangle$. Thus, to a good approximation, $|\langle AB\rangle| \ll K$. But since A and B are jointly Gaussian from Eq. (11), it then follows that A and B are uncorrelated Gaussian random processes.

The joint probability density of Q and α given by Eq. (11) is therefore

$$P(Q,\alpha)=\frac{1}{2\pi K}\left|\frac{dAdB}{dQd\alpha}\right| e^{-Q^2/2K}=\frac{2}{2\pi K}e^{-Q^2/2K} . \tag{19}$$

We see that α is uniformly distributed over the range 0 to 2π, independently of Q. Since β is similarly distributed, and α and β are independent, it follows that $\delta\equiv\alpha-\beta$ is also uniformly distributed over 0 to 2π, and the probability density $p(E)$ of $E=Q\cos\delta$ must be similar to the probability density of $A=Q\cos\alpha$. We have therefore shown that

$$p(E)=(2\pi K)^{-\frac{1}{2}}e^{-Q^2/2K} , \tag{20}$$

and the scattered field fluctuates in a Gaussian manner at each space-time point. We shall see, however, that the *random process* represented by the scattered field $\vec{E}(\vec{X},t)$ or $\vec{V}(\vec{x},t)$ is not Gaussian.

4. AMPLITUDE CORRELATIONS

Let us now form the second-order correlation function of the scattered field. From Eq. (9) we find

$$\Gamma^{(1,1)}(\vec{X}_1,\vec{X}_2,\tau)\equiv\langle\vec{V}^*(\vec{X}_1,t)\cdot\vec{V}(\vec{X}_2,t+\tau)\rangle=I_0k_0^4\langle\exp i[\beta(\vec{x}_0,t+\tau-R_{02}/c)-\beta(\vec{x}_0,t-R_{01}/c)]\rangle\exp(-ick_0\tau)$$
$$\times\iint_{\mathcal{V}}d^3x_1d^3x_2(R_1R_2)^{-1}\langle\Delta\chi(\vec{x}_1,t-R_1/c)\Delta\chi(\vec{x}_2,t+\tau-R_2/c)\rangle\exp i[\vec{k}_0\cdot(\vec{x}_2-\vec{x})+k_0(R_2-R_1)] , \tag{21}$$

where $R_1\equiv|\vec{X}_1-\vec{x}_1|$, $R_2\equiv|\vec{X}_2-\vec{x}_2|$, $R_{01}\equiv|\vec{X}_1-\vec{x}_0|$, $R_{02}\equiv|\vec{X}_2-\vec{x}_0|$, and we have taken the fluctuations of $\beta(\vec{x},t)$ and $\chi(\vec{x},t)$ to be statistically independent, as before. For the correlation of the susceptibility fluctuations we may use Eq. (13) to write

$$\langle\Delta\chi(\vec{x}_1,t-R_1/c)\Delta\chi(\vec{x}_2,t+\tau-R_2/c)\rangle=\mu(\vec{x}_2-\vec{x}_1,\tau+R_1/c-R_2/c)\approx\mu(\vec{x}_2-\vec{x}_1,\tau) , \tag{22}$$

since $\mu(\vec{x},\tau)$ does not vary appreciably over a time interval $(R_1-R_2)/c$. Similarly, if $|R_{01}-R_{02}|/c$ is much less than the coherence time of the incident light beam, as is almost certainly the case in practice, then, since $\beta(\vec{x},t)$ is a slowly varying function of time, we may write

$$\langle\exp i[\beta(\vec{x}_0,t+\tau-R_{02}/c)-\beta(\vec{x}_0,t-R_{01}/c)]\rangle\exp(-ick_0\tau)$$
$$=\langle\exp i[\beta(\vec{x}_0,t+\tau)-\beta(\vec{x}_0,t)]\rangle\exp(-ick_0\tau)=\gamma_0(\tau)\ \text{[by Eq. (8)]} , \tag{23}$$

where $\gamma_0(\tau)\equiv\langle\vec{V}_0^*(\vec{x}_0,t)\cdot\vec{V}_0(\vec{x}_0,t+\tau)\rangle/I_0$ (24)

is the normalized amplitude autocorrelation function of the incident laser beam.[29]

In order to arrive at a plausible form for $\gamma_0(\tau)$ for a single-mode laser beam, we make the usual assumption that the phase function $\beta(\vec{x}_0,t)$ may be considered to perform a one-dimensional random walk in time.[31,32] The phase changes are thereby viewed as the result of a very large number of small independent perturbations. Although a proper derivation of the laser spectral density requires a quantum treatment, this simple model appears to be satisfactory both as regards the intrinsic limitations of the laser process, and the external disturbances acting on the laser cavity. It is convenient to write

$$\beta(\vec{x}_0,t)=\int_0^t f(\vec{x}_0,t')dt' , \tag{25}$$

where $f(\vec{x}_0,t)$ is some random frequency function of zero mean, which is assumed to have a Gaussian probability density and negligible time correlation, and plays the role of the "velocity" of a Brownian particle. It is convenient to put

$$\langle f(\vec{x}_0,t)f(\vec{x}_0,t+\tau)\rangle=D\delta(\tau) , \tag{26}$$

where D is a constant characteristic of the "diffusion" rate of the phase $\beta(\vec{x},t)$, with a magnitude of the order of the spectral width of the laser beam. Then $\gamma_0(\tau)$ is given by

$$\gamma_0(\tau)=\langle\exp[i\int_t^{t+\tau}f(\vec{x}_0,t')dt']\rangle\exp(-ick_0\tau) ,$$

in which the first factor is the characteristic functional of a δ-correlated Gaussian random process. We may therefore make use of the general properties of the Gaussian characteristic functional,[33] to obtain

$$\gamma_0(\tau)=\exp[-\tfrac{1}{2}|\iint_t^{t+\tau}\langle f(\vec{x}_0,t')f(\vec{x}_0,t'')\rangle dt'dt''|]\exp(-ick_0\tau)=\exp(-\tfrac{1}{2}D|\tau|)\exp(-ick_0\tau) . \tag{27}$$

From Eqs. (21), (22), (23), and (27) we then have, if R_1 and R_2 are much greater than the linear dimensions of $\mathcal{V}$,

$$\Gamma^{(1,1)}(\vec{X}_1, \vec{X}_2, \tau) = (I_0 k_0^4/R_{01}R_{02})\exp(-\tfrac{1}{2}D|\tau| - ick_0\tau) \times \iint_{\mathcal{V}} d^3x_1 d^3x_2 \mu(\vec{x}_2 - \vec{x}_1, \tau)\exp i[\vec{k}_0 \cdot (\vec{x}_2 - \vec{x}_1) + k_0(R_2 - R_1)] . \quad (28)$$

We now introduce the far-field approximation as before, which allows us to write [c.f. Eq. (14)]

$$R_1 - R_2 = |R_{01}\vec{\rho}_1 - (\vec{x}_1 - \vec{x}_0)| - |R_{02}\vec{\rho}_2 - (\vec{x}_2 - \vec{x}_0)| \approx R_{01} - R_{02} - \vec{\rho}_1 \cdot (\vec{x}_1 - \vec{x}_0) + \vec{\rho}_2 \cdot (\vec{x}_2 - \vec{x}_0) , \quad (29)$$

where $\vec{\rho}_1$ and $\vec{\rho}_2$ are unit vectors in the directions $\vec{X}_1 - \vec{x}_0$ and $\vec{X}_2 - \vec{x}_0$. From Eqs. (28) and (29) we have

$$\Gamma^{(1,1)}(\vec{X}_1, \vec{X}_2, \tau) = (I_0 k_0^4/R_{01}R_{02})\exp(-\tfrac{1}{2}D|\tau|)\exp[-ik_0(c\tau + R_{01} - R_{02}) + i(\vec{K}_2 - \vec{K}_1)\cdot\vec{x}_0] \times \iint_{\mathcal{V}} d^3x_1 d^3x_2 \mu(\vec{x}_2 - \vec{x}_1, \tau)\exp i[\vec{k}_0 \cdot (\vec{x}_2 - \vec{x}_1) + \vec{K}_1 \cdot \vec{x}_1 - \vec{K}_2 \cdot \vec{x}_2] , \quad (30)$$

where $\vec{K}_1 = k_0\vec{\rho}_1$ and $\vec{K}_2 = k_0\vec{\rho}_2$ are the wave vectors of the scattered field in the measured directions. If the linear dimensions l_1, l_2, l_3 of $\mathcal{V}$ are great compared with the spatial correlation range of $\mu(\vec{x}, \tau)$ then, with the help of the substitutions $\vec{x}_2 - \vec{x}_1 = \vec{x}'$, $\vec{x}_2 + \vec{x}_1 = \vec{x}''$, this reduces to

$$\begin{aligned}\Gamma^{(1,1)}(\vec{X}_1, \vec{X}_2, \tau) &= (I_0 k_0^4/R_{01}R_{02})\exp(-\tfrac{1}{2}D|\tau|)\exp[-ik_0(c\tau + R_{01} - R_{02})] \\ &\quad \times \int d^3x' \mu(\vec{x}', \tau)\exp i[\vec{k}_0 - \tfrac{1}{2}(\vec{K}_1 + \vec{K}_2)]\cdot\vec{x}' \tfrac{1}{8}\int_{-l_1}^{l_1}\int_{-l_2}^{l_2}\int_{-l_3}^{l_3} d^3x'' \exp[\tfrac{1}{2}i(\vec{K}_2 - \vec{K}_1)\cdot\vec{x}''] \\ &= (I_0 k_0^4 \mathcal{V}/R_{01}R_{02})\exp(-\tfrac{1}{2}D|\tau|)\exp[-ik_0(c\tau + R_{01} - R_{02})] \; M[\vec{k}_0 - \tfrac{1}{2}(\vec{K}_1 + \vec{K}_2), \tau] \\ &\quad \times \prod_{j=1,2,3} \sin[\tfrac{1}{2}(\vec{K}_2 - \vec{K}_1)_j l_j]/\tfrac{1}{2}(\vec{K}_2 - \vec{K}_1)_j l_j , \end{aligned} \quad (31)$$

where $M(\vec{k}, \tau) = \int d^3x \mu(\vec{x}, \tau) e^{i\vec{k}\cdot\vec{x}}$ (32)

is the spatial Fourier transform of the susceptibility correlation function. Thus, from an exploration of the scattered field in various directions, it should be possible to determine the correlation properties of the scattering fluid.

A number of comments are perhaps worth making. For a fixed $\tfrac{1}{2}(\vec{K}_1 + \vec{K}_2)$ vector, the dependence of the correlation $\Gamma^{(1,1)}(\vec{X}_1, \vec{X}_2, \tau)$ on $\vec{K}_2 - \vec{K}_1$, i.e., on the separation of the points $\vec{X}_1$, $\vec{X}_2$, is governed entirely by the dimensions l_1, l_2, l_3 of the scattering region, and not at all by the correlation range of the elementary scatterers. No useful information about the properties of the scatterers can therefore be obtained from measurements of the spatial coherence at two points in the scattered field, which cannot equally (and more conveniently) be obtained from measurements at one point.[25]

If we allow the points $\vec{X}_1$ and $\vec{X}_2$ to coincide at $\vec{X}$, we obtain the somewhat simpler formula

$$\Gamma^{(1,1)}(\vec{X}, \vec{X}, \tau) = (I_0 k_0^4 \mathcal{V}/R_0^2)\exp(-\tfrac{1}{2}D|\tau| - ick_0\tau)M(\vec{k}_0 - \vec{K}, \tau) , \quad (33)$$

from which it is seen that the τ-dependence of $\Gamma^{(1,1)}(\vec{X}, \vec{X}, \tau)$ is a reflection of the spectral width both of the incident laser beam and of the susceptibility fluctuations. Whichever spectral width is the greater will tend to dominate the behavior in Eq. (33). Since the frequency ranges of typical fluid fluctuations appear to be of order kc/sec or tens of kc/sec,[2-9,1,16] while the spectral width D of even the most stable commercial single-mode laser is usually some hundreds of kc/sec, it is the $\exp(-\tfrac{1}{2}D|\tau|)$ term which is likely to determine the τ dependence of $|\Gamma^{(1,1)}(\vec{X}, \vec{X}, \tau)|$. We see therefore that the spectral width of the incident field cannot be neglected in the calculation of the correlation function $\Gamma^{(1,1)}$.

As a special case of Eq. (33), on putting $\tau = 0$, we obtain the mean scattered light intensity at $\vec{X}$,

$$\langle I(\vec{X}, t)\rangle = (I_0 k_0^4 \mathcal{V}/R_0^2)M(\vec{k}_0 - \vec{K}, 0) , \quad (34)$$

which also contains the information about the spatial correlations of the scatterers.

5. INTENSITY CORRELATIONS

Since it has become customary to study the properties of the scattered field with a combination of photodetector and spectral analyzer, and since the output of the spectral analyzer contains a contribution depending on the intensity autocorrelation function of the field[34] (in addition to the shot noise), we now examine the nature of this correlation. From Eq. (9) we can readily form the fourth-order correlation function defined by

$$\langle I(\vec{X},t)I(\vec{X},t+\tau)\rangle \equiv \langle \vec{V}^*(\vec{X},t)\cdot\vec{V}(\vec{X},t)\ \ \vec{V}^*(\vec{X},t+\tau)\cdot\vec{V}(\vec{X},t+\tau)\rangle$$

$$= I_0^2k_0^8 \int\int\int\int_{\mathcal{V}} (d^3x)^4\ (R_1R_2R_3R_4)^{-1}\langle \Delta\chi(\vec{x}_1, t-R_1/c)\Delta\chi(\vec{x}_2, t-R_2/c)$$

$$\times\,\Delta\chi(\vec{x}_3, t+\tau-R_3/c)\Delta\chi(\vec{x}_4, t+\tau-R_4/c)\rangle$$

$$\times\exp[i\vec{k}_0\cdot(-\vec{x}_1+\vec{x}_2-\vec{x}_3+\vec{x}_4)-ik_0(-R_1+R_2-R_3+R_4)]\,. \tag{35}$$

Since $\Delta\chi(\vec{x},t)$ is assumed to be a Gaussian random process, we may make use of the moment theorem for the Gaussian process to reduce the fourth-order correlation of $\Delta\chi$ to products of second-order correlations.[35] Thus,

$$\langle\Delta\chi(\vec{x}_1, t-R_1/c)\Delta\chi(\vec{x}_2, t-R_2/c)\Delta\chi(\vec{x}_3, t+\tau-R_3/c)\Delta\chi(\vec{x}_4, t+\tau-R_4/c)\rangle$$

$$=\mu(\vec{x}_2-\vec{x}_1, R_1/c-R_2/c)\mu(\vec{x}_4-\vec{x}_3, R_3/c-R_4/c)+\mu(\vec{x}_3-\vec{x}_1, \tau+R_1/c-R_3/c)\mu(\vec{x}_4-\vec{x}_2, \tau+R_2/c-R_4/c)$$

$$+\mu(\vec{x}_4-\vec{x}_1, \tau+R_1/c-R_4/c)\mu(\vec{x}_3-\vec{x}_2, \tau+R_2/c-R_3/c)$$

$$=\mu(\vec{x}_2-\vec{x}_1, 0)\mu(\vec{x}_4-\vec{x}_3, 0)+\mu(\vec{x}_3-\vec{x}_1, \tau)\mu(\vec{x}_4-\vec{x}_2, \tau)+\mu(\vec{x}_4-\vec{x}_1, \tau)\mu(\vec{x}_3-\vec{x}_2, \tau), \tag{36}$$

where the last equation follows from the previous one [c.f. Eq. (14)] by virtue of the fact that $\mu(\vec{x},\tau)$ is a slowly varying function of τ. If $\vec{X}$ is sufficiently distant from the scatterer so that the factor $(R_1R_2R_3R_4)^{-1}$ may be replaced by $1/R_0^4$ ($R_0=|\vec{X}-\vec{x}_0|$) under the integral, and the far-field approximation given by Eq. (29) may be used, and if we assume, as before, that the linear dimensions l_1, l_2, l_3 of $\mathcal{V}$ are all large compared with the correlation range of $\mu(\vec{x},\tau)$, then with the help of Eq. (36), Eq. (35) reduces to

$$\langle I(\vec{X},t)I(\vec{X},t+\tau)\rangle = (I_0^2k_0^8\mathcal{V}^2/R_0^4)\int\int d^3x'd^3x''\mu(\vec{x}',0)\mu(\vec{x}'',0)\exp[i(\vec{k}_0-\vec{K})\cdot(\vec{x}'+\vec{x}'')]$$

$$+(I_0^2k_0^8/R_0^4)\int\int d^3x'd^3x''\mu(\vec{x}',\tau)\mu(\vec{x}'',\tau)\exp[i(\vec{k}_0-\vec{K})\cdot(\vec{x}''-\vec{x}')]$$

$$\times\int\int_{\mathcal{V}} d^3x_1d^3x_2\exp[2i(\vec{k}_0-\vec{K})\cdot(\vec{x}_2-\vec{x}_1)]$$

$$+(I_0^2k_0^8\mathcal{V}^2/R_0^4)\int\int d^3x'd^3x''\,\mu(\vec{x}',\tau)\,\mu(\vec{x}'',\tau)\,\exp i(\vec{k}_0-\vec{K})\cdot(\vec{x}'-\vec{x}'')$$

$$=\frac{I_0^2k_0^8\mathcal{V}^2}{R_0^4}\left\{M^2(\vec{k}_0-\vec{K},0)+|M(\vec{k}_0-\vec{K},\tau)|^2\left[1+\prod_{j=1,2,3}\left(\frac{\sin(\vec{k}_0-\vec{K})_jl_j}{(\vec{k}_0-\vec{K})_jl_j}\right)^2\right]\right\}\,. \tag{37}$$

If the linear dimensions of the scattering region are very great compared with the wavelength of the incident light, then the third term will be negligible compared with the second for almost all directions of scattering except the forward direction. On subtracting $\langle I(\vec{X},t)\rangle^2$ given by Eq. (34) from both sides of Eq. (37), we obtain

$$\langle\Delta I(\vec{X},t)\Delta I(\vec{X},t+\tau)\rangle=\frac{I_0^2k_0^8\mathcal{V}^2}{R_0^4}|M(\vec{k}_0-\vec{K},\tau)|^2\left\{1+\prod_{j=1,2,3}\left[\frac{\sin(\vec{k}_0-\vec{K})_jl_j}{(\vec{k}_0-\vec{K})_jl_j}\right]^2\right\}\,. \tag{38}$$

We have shown, therefore, that the properties of the susceptibility correlations are simply related to the intensity correlations of the scattered field. They can therefore be obtained from an autocorrelation or spectral analysis of the current fluctuations of a photodetector exposed to the scattered field. Thus, it is now well known that the spectral density $G(\omega)$ of the current fluctuations of an illuminated photodetector is given by[34]

$$G(\omega)=\alpha cS\langle I(\vec{X},t)\rangle\,|K(\omega)|^2[1+\alpha cS\,\Psi(\vec{X},\omega)/\langle I(\vec{X},t)\rangle\,]\,, \tag{39}$$

where α is the quantum efficiency of the photodetector and S its surface area, $\Psi(\vec{X},\omega)$ is the Fourier transform of $\langle\Delta I(\vec{X},t)\Delta I(\vec{X},t+\tau)\rangle$ and $K(\omega)$ is the frequency response of the detector circuit. Alternatively, the intensity correlation on the left of Eq. (38) could be obtained from an analysis of the emission times of photoelectric pulses at the photodetector.[34] But perhaps the most interesting feature of Eq. (38) is the fact that the correlation properties of the incident laser field do not appear in it. The spectral linewidth of the laser beam therefore plays no role at all in the measured photoelectric fluctuations. We emphasize once again that this linewidth is not negligible compared with the spectral width of the susceptibility fluctuations; on the contrary, it is likely to be many times as great. It is because of the constant absolute amplitude of the field from a single-mode laser, that this linewidth does not appear in Eq. (38). However, with a scatterer of sufficiently large dimensions l_1, l_2, l_3 this conclusion would not hold, and the phase fluctuations of the laser beam would show up in the intensity correlations of the scattered light.

6. DISCUSSION

From Eqs. (38) and (33) we find that, for any direction other than the forward direction,

$$\langle \Delta I(\vec{X}, t)\, \Delta I(\vec{X}, t+\tau)\rangle = |\Gamma^{(1,1)}(\vec{X}, \vec{X}, \tau)|^2 e^{D|\tau|}, \qquad (40)$$

so that the complex scalar amplitude $V(\vec{X}, t)$ of the scattered field does not satisfy the moment theorem for a Gaussian random process. The scattered field is therefore not a Gaussian field, i.e. it is not a field obeying the usual statistics of thermal light, despite the fact that the instantaneous wave amplitude at each space-time point has a Gaussian probability density, as we showed in Section 3. Evidently the non-Gaussian features of the field will only become apparent if we examine the joint probabilities of wave amplitudes separated by time intervals of order $1/D$ at the same point (or corresponding intervals at different points). But, clearly, the non-Gaussian properties will not appear at all in measurements involving only the correlations of the light intensity.

By starting from Eq. (9) we may readily show that the nth moment of $I(\vec{X}, t)$ satisfies the relation

$$\langle I(\vec{X}, t)^n\rangle = n!\,\langle I(\vec{X}, t)\rangle^n, \qquad (41)$$

at a distant point $\vec{X}$, other than in the forward direction, so that $I(\vec{X}, t)$ has an exponential probability density. As a result, the number of photoelectric counts registered by a photodetector at $\vec{X}$ in a short time interval will obey the Bose-Einstein distribution,[34,36] which is characteristic of thermal light. However, it should now be clear that it is not possible to draw the conclusion that the scattered field is a Gaussian field (in the sense that the probability functional or phase-space functional is Gaussian) on the basis of photoelectric counting measurements, as has sometimes been done.[8]

As we pointed out in the introduction, it has been customary in many previous treatments of the scattering problem to treat the incident laser field as strictly monochromatic, and to take it for granted that the scattered field is a Gaussian field. Inspection of Eq. (40) shows that, if we put $D = 0$, then the Gaussian condition does indeed hold. In other words, the effect introduced in Eq. (33) by the neglect of the laser spectral width, and the effect introduced in Eq. (40) by the Gaussian assumption, cancel each other, and the resulting equation for the intensity correlation is then correct. But this fortunate circumstance, which depends on the special properties of a single-mode laser beam, must be regarded as somewhat fortuitous.

It is not difficult to envisage a situation in which neglect of the laser linewidth would not give the right answer. Let us consider a laser which is oscillating in two modes simultaneously, and let us suppose that these modes are centered on frequencies k_1 and k_2 [with $k_0 = \frac{1}{2}(k_1 + k_2)$], that they have associated linewidths D_1 and D_2, and equal constant amplitudes $(\frac{1}{2}I_0)^{1/2}$. For simplicity we assume that the two modes are statistically independent. Then, if these modes result in scattered complex wave amplitudes $\vec{V}_1(\vec{X}, t)$ and $\vec{V}_2(\vec{X}, t)$ respectively, the resultant complex wave amplitude at $(\vec{X}, t)$ will be given by

$$V(\vec{X}, t) = \vec{V}_1(\vec{X}, t) + \vec{V}_2(\vec{X}, t), \qquad (42)$$

so that

$$I(\vec{X}, t) = I_1(\vec{X}, t) + I_2(\vec{X}, t) + \vec{V}_1^*(\vec{X}, t)\cdot\vec{V}_2(\vec{X}, t) + \text{c. c.}$$

and

$$\langle I(\vec{X}, t) I(\vec{X}, t+\tau)\rangle = \langle I_1(\vec{X}, t) I_1(\vec{X}, t+\tau)\rangle + \langle I_2(\vec{X}, t) I_2(\vec{X}, t+\tau)\rangle + 2\langle I_1(\vec{X}, t)\rangle\langle I_2(\vec{X}, t)\rangle + \langle \vec{V}_1^*(\vec{X}, t)\cdot\vec{V}_1(\vec{X}, t+\tau)\rangle\langle \vec{V}_2(\vec{X}, t)\cdot\vec{V}_2^*(\vec{X}, t+\tau)\rangle, \qquad (43)$$

provided both modes are similarly polarized. With the help of Eqs. (33), (34), and (38), we can evaluate each term in this equation, and we find that, except for points in the forward direction,

$$\langle \Delta I(\vec{X}, t)\, \Delta I(\vec{X}, t+\tau)\rangle = (I_0^2 k_0^8 \mathfrak{v}^2 / R_0^4)$$

$$\times |M(\vec{k}_0 - \vec{K}, \tau)|^2 \tfrac{1}{2}\{1 + \cos[(k_1 - k_2)\tau] \times \exp[-\tfrac{1}{2}(D_1 + D_2)|\tau|]\}, \quad (44)$$

which should be compared with Eq. (38) under the same conditions. This time we note that the spectral features of the laser beam appear explicitly in the second term in Eq. (44). Whether the contribution of the second term is apparent in measurements of the spectral density $G(\omega)$ of the photoelectric current fluctuations (given by Eq. (39)), depends largely on the mode separation $k_1 - k_2$. If this frequency difference is appreciably less than about a Mc/sec, as it might be for off-axis modes, the effect of the second term may well be to distort the measured spectral density. This situation is illustrated in Fig. 1, in which the Fourier transform $\Psi(\omega)$ of

$$\langle \Delta I(\vec{X}, t)\, \Delta I(\vec{X}, t+\tau)\rangle$$

is shown as a function of frequency ω. If $D_1 + D_2$ is comparable with – or greater than – $|k_1 - k_2|$, the effect of light beating between the modes will be superimposed on the contribution from the susceptibility fluctuations. It is possible that this phenomenon may have contributed to a few rather wide measured spectral densities which have been reported.

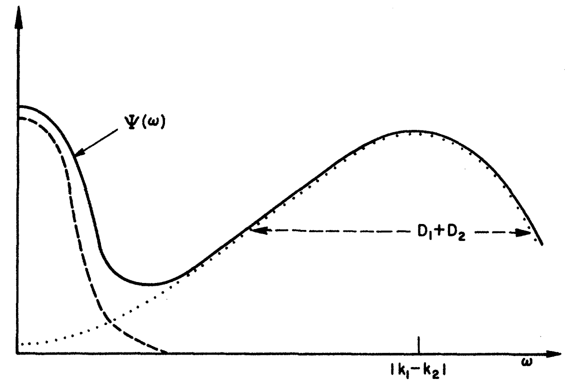

FIG. 1. Illustrating the form of the spectral density $\Psi(\omega)$ of the intensity fluctuations of scattered light, for a two-mode laser beam. The broken and dotted curves show the separate contributions of the first and second terms in Eq. (44), respectively.

We conclude, therefore, that, while the effect of the laser spectral width is unimportant in the usual measurements so long as a single-mode laser is used as source, this is not necessarily so in all measurements, or under multimode conditions.

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Perturbation Study of Some Excited States of Two-Electron Atoms

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A perturbation study of the NP states of two-electron atoms has been made. In particular, oscillator strength values for the (1^1S, 2^1P) and (2^3S, 2^3P) transitions are obtained. The 2^1P and 2^3P states are studied through ninth and tenth order, respectively. In addition, the N^1P and N^3P states are studied in first order through the $10P$ member of the series. Perturbation energy coefficients and other expectation values for several important operators have been computed. Perturbation energy coefficients for the 1^1S state (through 25th order) and 2^3S state (through 17th order) are also reported. Where comparison is possible, these results are in satisfactory agreement with the results obtained from variational calculations by C. L. Pekeris and co-workers. The variational-perturbation method for excited states requires auxiliary conditions on the perturbation wave functions. The condition on the nth-order wave function is derived here. This is a generalization of the first-order condition given by Sinanoğlu.

I. INTRODUCTION AND PROCEDURE

This study was undertaken in order to apply the variational-perturbation methods previously developed[1] to a study of the oscillator strengths in the 2^1P to 1^1S and the 2^3P to 2^3S transitions for the helium isoelectronic series. This task necessitated the construction of accurate $2P$ perturbation wavefunctions through high orders, and, concomitantly, the availability of 1^1S and 2^3S wave functions of comparable accuracy and order. For reasons of computational convenience, a new 1^1S ground-state wave function was determined, although similar wave functions already are in existence. In addition the $2P$ perturbation wave functions are themselves of interest, and a study has been made of their eigenvalues, expectation values with certain operators, etc. The perturbation energy coefficients for the 1^1S and 2^3S states are also reported. The S state calculations were regarded as of secondary interest, and no detailed study of them is presented. Further, first-order studies of the NP states, N from 3 to 10, were completed, and are briefly reported.

1. Notation

The notation used here is as follows. Let the Hamiltonian be given in atomic units[2] by

$$H = H_0 + H_1 = H_0 + 1/Zr_{12}, \quad (1)$$

where Z is the nuclear charge and $1/Zr_{12}$ is regarded as the perturbation. Then a solution $\Psi^{(M)}$ for the Mth state can be written

$$\Psi^{(M)} = \sum_n Z^{-n} \psi_n^{(M)}, \quad (2)$$

with eigenvalue