Quantum theory of dynamic multiple light scattering in fluctuating disordered media

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We formulate a quantum theory of dynamic multiple light scattering in fluctuating disordered media and calculate the fluctuation and the autocorrelation function of the photon number operator for light transmitted through a disordered slab. The effect of disorder on the information capacity of a quantum communication channel operating in a disordered environment is estimated, and the use of squeezed light in diffusing-wave spectroscopy is discussed.

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Many disordered media (suspensions, emulsions, foams, semiconductor powders, biological tissues, etc.) scatter light diffusely and appear opaque. Statistical properties of light scattered from disordered media are of fundamental importance for such applications as wireless communications [1–3] and different types of diffuse optical spectroscopies (diffuse transmission spectroscopy [4], diffusing-wave spectroscopy [5,6], etc.). With a few notable exceptions [7,8], most theoretical studies in this field of research have been conducted by treating light as a classical wave [9,10], without accounting for the quantum nature of light. Meanwhile, recent experiments demonstrate that quantum properties of light in disordered media (such as quantum noise [11] and photon statistics [12]) can be of interest from both fundamental and applied points of view.

The purpose of the present paper is to fill up an important gap in the quantum theory of multiple light scattering by developing a quantum description of dynamic multiple light scattering in fluctuating disordered media. In contrast to the previous work [7,8], we take into account the evolution of a disordered medium (e.g., Brownian or more complex motion of scattering centers) in the course of a multiple scattering experiment. This leads to an interesting and rich problem of quantum dynamics, where the quantum fluctuations of light are coupled with the classical fluctuations of the medium. Our results allow us to estimate the impact of disorder on the information capacity of quantum communication channels, and to lay a foundation for the use of nonclassical light as a probe of disordered media.

We consider a slab of disordered medium that, for concreteness, we assume to be a suspension of mobile, elastically scattering particles (see Fig. 1). For each frequency \( \omega \), light incident on the slab can be decomposed into \( M \) transverse modes that we denote by indices \( a' \) and \( b' \) for modes on the left and on the right of the slab, respectively. All but one of the incoming modes are assumed to be in vacuum states. The state of the mode \( a' = a \) on the left can be arbitrary, and it corresponds to the light that one sends into the medium in an experiment. We are interested in the properties of multiply scattered light in the transmitted, outgoing mode \( b \).

To quantize the electromagnetic field we use the same method as Beenakker et al. [7] and Lodahl et al. [8], who considered disordered media with immobile scattering centers. An annihilation operator \( \hat{a}(\omega) \) is associated with each mode, and standard bosonic commutation relations are assumed [7,8]:

\[ [\hat{a}(\omega), \hat{a}^\dagger(\omega')] = \delta_{ij} \delta(\omega - \omega'), \]

where \( i \) and \( j \) run either over all incoming modes or over all outgoing modes. We now note that the fluctuating disordered medium acts as a beam splitter having \( M \) input and \( M \) output ports and time-dependent transmission and reflection coefficients \( \tau_{a'b}(t) \) and \( \rho_{a'b}(t) \). By analogy with the work of Hussein et al. [13], who dealt with a “regular” beam splitter (\( M = 2 \)) having time-dependent properties, we assume that \( \tau_{a'b}(t) \) and \( \rho_{a'b}(t) \) are frequency independent and that they do not vary significantly during the typical time it takes for light to cross the disordered sample. The former assumption requires a sufficiently narrow bandwidth \( B \) of the incident light, whereas the latter is justified provided that the dynamics of the medium is slow. With these assumptions in hand, the input-output relations for \( \hat{a}(\omega) \) read

\[ \hat{a}(t) = \sum_{a'} \tau_{a'b}(t) \hat{a}_{a'}(t) + \sum_{b'} \rho_{a'b}(t) \hat{a}_{b'}(t), \]

where \( \hat{a}_{a'}(t) \) denotes the Fourier transform of \( \hat{a}(\omega) \). The use of \( \hat{a}_b(t) \) is permissible provided that \( B \) is much smaller than

FIG. 1. Ensemble of mobile scattering particles (circles) is illuminated from the left by a wave in the incoming mode \( a \). All other incoming modes are in vacuum states (modes \( a' \) on the left and \( b' \) on the right). We are interested in the fluctuations of transmitted light in the outgoing mode \( b \). Dotted line illustrates the multiple scattering with a mean free path \( l \approx L \) that light undergoes inside the medium.
the central frequency $\omega_0$ of the incident light [13,14]. Similarly to the case of immobile scatterers [7,8], Eq. (2) conserves the commutation relations (1), due to the unitarity of the scattering matrix composed of $\tau_{\alpha\beta}(t)$ and $\rho_{\alpha\beta}(t)$ in the absence of absorption. However, in contrast to the previous work [7,8], Eq. (2) implies the possibility of energy exchange between different spectral components upon transmission of light through a fluctuating disordered medium. This becomes clear when we back-Fourier-transform Eq. (2), because products of time-dependent quantities become convolutions in the frequency space. Such an energy exchange is by no means a nonlinear phenomenon, but is a manifestation of the Doppler effect due to scattering of light on moving particles.

To establish a link between $\hat{a}(t)$ and measurable quantities, we define a flux operator $\hat{I}(t)=\hat{a}^\dagger(t)\hat{a}(t)$ that, when integrated from $t$ to $t+T$, yields a photon number operator $\hat{n}_i(t,T)$ corresponding to the number of photocounts measured by an ideal, fast photodetector illuminated by light in the mode $i$ during some sampling time $T$ [14]. A relation between the average values of operators $\bar{n}_i(t,T)$ and $\hat{n}_i(t,T)$ corresponding to the outgoing mode $b$ and the incoming mode $a$, respectively, is readily found from Eqs. (1) and (2): $\bar{n}_b=\bar{I}_{ab}(\bar{n}_a)$. Here $T_{ab}(t)|=\langle T_{ab}(t)\rangle^2$, the angular brackets denote the quantum-mechanical expectation value, and the overbar denotes averaging over disorder. By the ergodicity hypothesis, the simultaneous averaging $(\bar{\cdots})$ over both disorder and quantum fluctuations can be replaced by time averaging in a realistic experiment with a fluctuating disordered medium, provided that the incident light is stationary. On the contrary, there is no simple way of obtaining averages over either disorder or quantum fluctuations separately.

Although the relation between $\langle \hat{n}_b \rangle$ and $\langle \hat{n}_a \rangle$ is independent of the quantum state of the incident light, the latter is crucial for the second-order statistics of the photon number operator. To illustrate this, we consider a particularly simple case of a stationary, quasimonochromatic incident beam for which both $(\hat{I}_a(t))$ and $(\hat{I}_b(t))$, with $\cdots$ denoting normal ordering [15], can be considered time independent on the scale of $T$. This includes a number of practically important examples on which we will dwell in more detail: coherent light, continuous-wave quadrature squeezed light with a rectangular squeezing spectrum of width $B\ll 1/T$ around the central frequency of a perfectly monochromatic mean field $A_0 \exp(-i\omega_0 t+id)$ [16], light in a generalized continuum number state $|n,\xi\rangle$ with a box-shaped envelope function $\xi(t)=(t_{\text{max}}-t_{\text{min}})^{-1/2} \exp(-i\omega_0 t)$, where $t_{\text{min}}\leqslant t \leqslant t_{\text{max}}$ and chaotic light with a long coherence time $t_{\text{coh}}\gg T$. Making use of Eqs. (1) and (2), we find that the normalized variance of photon number fluctuations in the outgoing mode $b$ can be written as

$$\delta_b^2=\Var[\hat{n}_b]/\langle \hat{n}_b \rangle^2=1+\delta_{\text{class}}^b+Q_a/\langle \hat{n}_a \rangle(1+\delta_{\text{class}}^a).$$

Here $\Var[\hat{n}_a]/\langle \hat{n}_a \rangle^2$ is the average photon number, and $Q_a=\Var[\hat{n}_a]/\langle \hat{n}_a \rangle-1$ is the Mandel parameter [15], corresponding to the incident light beam. $\delta_{\text{class}}^a$ is the normalized variance of time-integrated intensity in the mode $a$ that obtains from Eq. (2) by ignoring the quantum nature of light and dropping the operator carets of $\hat{a}(t)$. $\langle \hat{n}_b \rangle=\langle \hat{n}_a \rangle T_{ab}(t)|=\langle T_{ab}(t)\rangle t_{\text{coh}}/T$. Simultaneously, $\langle \hat{n}_a \rangle$ (which both $\delta_{\text{class}}^a$ and $\delta_{\text{class}}^b$ cannot be described classically and to which Mandel’s formula [17] corresponds) is readily found from Eqs. (1) and (2).

The mere fact that Eq. (3) contains both classical (or “wave”) and quantum (or “particle”) contributions is not surprising, and could be anticipated already from early ideas of Einstein (the so-called Einstein fluctuation formula) [17,18]. However, the exact structure of Eq. (3) and, in particular, the way it depends on the quantum state of the incident light (i.e., on the Mandel parameter $Q_a$) is difficult to conjecture without an explicit calculation. The main conclusion following from Eq. (3) is that, in the general case, the variance of photon number fluctuations in transmission through a fluctuating disordered medium is not simply a sum of quantum and classical (due to disorder) contributions, but rather a much more complicated object. In particular, the last term of Eq. (3) mixes classical fluctuations and quantum noise in a multiplicative way. Only for incident light in the coherent state does $Q_a=0$ and Eq. (3) reduces to a sum of shot noise $1/\langle \hat{n}_b \rangle$ and classical noise $\delta_{\text{class}}^b$. This is the minimum value of $\delta_b^2$ that can be obtained for light in a state admitting classical description. An “excess noise” $\delta_{\text{excess}}^2$—defined as the difference between $\delta_b^2$ and $1/\langle \hat{n}_b \rangle+\delta_{\text{class}}^b$—arises for other states. For chaotic light, for example, $Q_a=\langle \hat{n}_a \rangle$ and $\delta_{\text{excess}}^2=1+\delta_{\text{class}}^a$. Note that, for both coherent and chaotic states, we do not really need the quantum model developed here, but can instead use the semiclassical Mandel formula [15] as was done in Ref. [12].

We now turn to nonclassical light—light in states that cannot be described classically and to which Mandel’s formula does not apply. For incident light in the number state $|n_a,\xi\rangle$, $Q_a=-1$, and the excess noise is negative: the fluctuations of transmitted photon number are suppressed below the simple sum of shot noise $1/\langle \hat{n}_b \rangle$ and classical fluctuation $\delta_{\text{class}}^b$. $\delta_b^2$ becomes even less than the bare shot noise $1/\langle \hat{n}_b \rangle$ if $n_a<1+\delta_{\text{class}}^a$. An interesting situation occurs for the single-photon-number state $(n_a=1)$: the relative fluctuation of transmitted photon number becomes independent of $\delta_{\text{class}}^a$. Another important example of nonclassical light is the continuous-wave squeezed light for which $Q_a=(\langle \hat{n}_b \rangle)/(r,\phi,A_0^2/B,1)$ [16], where $r$ is the squeezing parameter and we define an auxiliary function

$$f(r,\phi,x,y)=[xy(e^{-2r} \cos^2 \phi + e^{2r} \sin^2 \phi-1) + y^2 \sin^2 r \cosh 2r]/(x+y)^2.$$

For the amplitude-squeezed light ($\phi=0$) the Mandel param-
FIG. 2. (Color online) Excess noise of photon number $\hat{n}_b$ in the transmitted mode $b$ for the incident light (mode $a$) in the continuous-wave amplitude-squeezed state with a narrow rectangular squeezing spectrum of width $B=1/T$. The excess noise is defined as the difference between the variance of photon number fluctuations $\bar{\delta}_b^2$, the shot noise $1/\langle \hat{n}_b \rangle$, and the classical noise $\delta^c_{\text{class}}$. It is shown as a function of sampling time $T$ and squeezing parameter $r$ for a suspension of small dielectric particles in Brownian motion (diffusion coefficient $D_b$). The unit of time is $t_0=\hbar^2/6\kappa^2D_b$, where $\hbar$ is the mean free path, $L\gg l$ is the sample thickness, and $k\gg l$ is the wave number of light. The characteristic decay time $t_0$ is set equal to $B$. Thick black lines show the excess noise as a function of sampling time for $r=0, 0.2, 0.4, 0.6, 0.8,$ and $1.0$.

er is negative if $r<r_0$, with $r_0$ depending on $A_0^2/B$, leading to $\delta^c_{\text{excess}}<0$, similarly to the number state. This is illustrated in Fig. 2 for $C_T^{ab}(t)=(t/t_0)\sinh^2t/t_0$, which corresponds to a suspension of small Brownian particles [5,6]. In the above expression for $C_T^{ab}(t)$, corrections of order $1/g$ (with $g\gg 1$ the dimensionless conductance of the disordered sample) are neglected. The characteristic decay time $t_0$ of $C_T^{ab}(t)$ is defined in the caption of Fig. 2. With $A_0^2/B=1$ chosen for this figure the excess noise is negative for $r<r_0\approx 0.6$.

It is instructive to compare Eq. (3) with the result obtained by Lodahl et al. for the variance of photon number fluctuations in a disordered medium with immobile scattering centers, Eq. (4a) of Ref. [8(a)]. The two results look quite different for two reasons. First, the authors of Ref. [8] made use of an explicit expression for the second moment of the transmission coefficient $T_{ab}$, which we do not need to detail and which, in principle, could be calculated with any desired degree of accuracy. Second, and most important, Lodahl et al. use a slightly different averaging procedure: they first calculate the quantum-mechanical expectation values $\langle \hat{n}_b \rangle$ and $\langle \hat{n}_b^2 \rangle$ and then average $\langle \hat{n}_b^2 \rangle - \langle \hat{n}_b \rangle^2$ over disorder, whereas we perform both quantum-mechanical and disorder averages simultaneously. As a result, the authors of Ref. [8] consider $\langle \hat{n}_b^2 \rangle - \langle \hat{n}_b \rangle^2$, whereas we deal with $\langle \hat{n}_b^2 \rangle - \langle \hat{n}_b \rangle^2$. The two ways of performing averaging correspond to two different experimental situations. In the static disordered medium considered by Lodahl et al., one could indeed imagine first performing the average over quantum fluctuations for a given (frozen) disorder and then averaging over disorder. By contrast, in the fluctuating medium considered in the present paper, quantum and disorder averages cannot be separated and have to be performed simultaneously (except, perhaps, in a hypothetical experiment involving a large number of disordered samples with microscopically identical dynamics). If, despite its irrelevance for realistic experiments with fluctuating disordered media, we calculate the same quantity as Lodahl et al. did, we obtain

$$\frac{\langle \hat{n}_b^2 \rangle - \langle \hat{n}_b \rangle^2}{\langle \hat{n}_b \rangle} = T_{ab} + T_{ab}^2 Q_a(1 + \delta_{\text{class}}^3).$$

(4)

This equation coincides with Eq. (4a) of Ref. [8] if we note that $Q_a=F_a-1$ (with $F_a$ the Fano factor) and set $\delta_{\text{class}}^3=1 + 8/3g$ which corresponds to a disordered medium with immobile scattering centers.

Equation (3) can be used to estimate the effect of disorder on the information capacity of quantum communication channels operating in disordered environments. As an example, let us consider a communication channel connecting Alice on the left of the disordered medium in Fig. 1 and Bob on the right. Alice communicates with Bob by sending a sequence of number states, each of duration $T$ and containing up to $n_a^{\max}$ photons, in the incoming mode $a$. Bob reconstructs the message of Alice by counting photons in the outgoing mode $b$ during the same time intervals $T$. By using the theory of linear bosonic communication channels [19] and Eq. (3), we find that the information capacity of such a channel is

$$C = \frac{n_a^{\max}T_{ab}}{Te \ln 2} \left[ 1 + \frac{1}{2}n_a^{\max}T_{ab} \right]$$
$$\times \left[ \frac{1}{n_a^{\max}} - \frac{1}{e} - \delta_{\text{class}}^3 \left( 1 - \frac{1}{n_a^{\max}} \right) \right] \text{ (bits/s)},$$

(5)

where we drop higher-order terms in $n_a^{\max}T_{ab}\ll 1$, which is justified by the condition $T_{ab}\ll 1$ typical for transmission of light through a diffusely scattering medium. As could be expected, the (classical) fluctuations $\delta_{\text{class}}^3$ reduce the capacity of the channel. However, this reduction is only second order in $n_a^{\max}T_{ab}\ll 1$. The drop of capacity in the presence of disorder is therefore mainly due to the reduced average value of the transmission coefficient $T_{ab}$ rather than to the fluctuations of the latter. Interestingly, $C$ appears to be independent of $\delta_{\text{class}}^3$ if $n_a^{\max}=1$ (Alice sends either one or no photons).

The condition $n_a^{\max}T_{ab}\ll 1$ used to derive Eq. (5) restricts the number of photons in individual number states that play the role of “letters” of which Alice composes her messages, but this condition restricts neither the total length of a message, nor the amount of information that it contains. In the opposite limit of $n_a^{\max}T_{ab}\gg 1$, quantum effects become negligible and one can apply the classical information theory [20]. In the intermediate regime of $n_a^{\max}T_{ab}\sim 1$, Eq. (3) is not sufficient for the calculation of capacity and knowledge of
higher moments of $\hat{n}_b$ is required. Study of these higher moments is beyond the scope of the present paper.

Correlation of photon numbers in the same mode $b$ but in two different, nonoverlapping time intervals,

$$C^n_b(t) = \frac{\langle \hat{n}_b(t_1,T)\hat{n}_b(t_1 + t,T) \rangle - 1}{\langle \hat{n}_b(t_1,T) \rangle \langle \hat{n}_b(t_1 + t,T) \rangle}, \quad (6)$$

is also affected by the quantum state of incident light and can be reduced to

$$C^n_b(t) = [1 + C^n_a(t)]C^ab_T(t) + C^n_a(t), \quad (7)$$

where $C^n_a(t)$ is the autocorrelation function of photon number fluctuations in the incident mode $a$. In order to put Eq. (7) in a simple form, the typical decay time of $C^ab_T(t)$ is assumed to be much longer than $T$. Equation (7) is more general than Eq. (3) and requires neither stationarity nor monochromaticity of the incident light. For light in the coherent state $C^n_a(t)=0$ and $C^ab_T(t)=C^ab_0$. This result actually lies at the heart of diffusing-wave spectroscopy (DWS) and other photon correlation techniques that identify the measured $C^n_b(t)$ with $C^ab_T(t)$ and interpret it in terms of classical wave scattering.

For nonclassical states of incident light $C^n_a(t)$ is not equal to $C^ab_T(t)$. For the number state $|n_a,\xi\rangle$, $C^n_a(t)=-1/n_a$ and $C^ab_T(t)$ can take negative values and becomes independent of time for $n_a=1$. These strange properties are direct consequences of the conservation of total number of photons—a conservation law which does not apply to the coherent state where the number of photons is not a good quantum number. For squeezed light we find $C^n_a(t)=f(r,\phi,\Delta^2/2B,\sin(c n B t))$. As illustrated in Fig. 3, squeezing induces oscillations of $C^n_a(t)$ and decreases its value at short $t$. The latter reaches a minimum at some $r=r_1$ ($r_1=0.3$ in Fig. 3) and then grows again, exceeding 1 at large $r$. The results presented in Fig. 3 open a way to performing DWS with squeezed light; the necessary experimental setup is completely analogous to the “traditional” DWS one [5,6], except that a source of squeezed light should be used instead of a laser. A completely new experiment that has no analog in the realm of classical light scattering would be performing DWS with squeezed vacuum (i.e., with incident light in a squeezed state with $A_0=0$).

Provided that $T_{ab}(t)$ is replaced by $T_a(t)=\Sigma_0 T_{ab}(t)$ in the definition of $C^ab_T(t)$, the main results of this paper—Eqs. (3) and (7)—also hold for the variance and the autocorrelation function of $\hat{n}=\Sigma_0 \hat{n}_b$—a photon number operator corresponding to the total transmission measurement. Equations (3) and (7) thus implicitly account for all possible long-range (in space or in time) correlations of $\hat{B}(t)$ that were extensively studied previously [9,10]. The effect of long-range correlations on photon number fluctuations in total transmission through a suspension of scattering particles in Brownian motion has been recently studied by Balog et al. [12] for incident light in a coherent state, and the result following for this case from Eq. (3) has been confirmed experimentally.

In conclusion, we developed a quantum description of dynamic multiple light scattering in fluctuating disordered media. We calculated the fluctuation and the autocorrelation function of the photon number operator in transmission through a slab of mobile scattering particles. We applied our results to estimate the effect of disorder on the information capacity of a quantum communication channel operating in a disordered environment and to discuss the possibilities of performing diffusing-wave spectroscopy with squeezed light. Our results suggest that using nonclassical light (single photons, light in number states, squeezed light) opens interesting perspectives in the optics of disordered media.

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