Investigation of the temporal fluctuations in dissipative quantum systems in connection with the experimental discovery of a new type of nonlinear scattering of light

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(Submitted 2 July 1986)
Zh. Eksp. Teor. Fiz. **93**, 54–64 (July 1987)

A quantum theory of fluctuations is constructed for dissipative systems that interact with the environment, and whose dynamics is therefore not described by a Hamiltonian only. The intensity and shape of the spectral line due to the scattering of light by the space-time fluctuations in the permittivity of the medium, fluctuations which arise as a result of the heating of the medium around the impurity molecules resonantly absorbing the incident light and transferring the absorbed energy to the medium, are computed.

1. INTRODUCTION

Belyaev *et al.*[1] investigated experimentally the scattering of $\lambda = 503$-nm light in water containing a small admixture of I_2 molecules, which absorbed this light. In the general case we can consider a medium containing a small admixture of resonance particles that absorb light passing through the medium; the medium itself does not absorb the light. In Ref. 1 they observed nonlinear scattering of light by the space-time fluctuations of the medium's permittivity that arise as a result of the heating of the medium around the impurity particles that resonantly absorb the incident light and transfer the absorbed energy to the medium. The temporal fluctuations of the temperature of the medium in such a physical situation are theoretically considered in Ref. 2 on the basis of the photon concept.

For the purpose of interpreting the experiment reported in Ref. 1, and determining the limits of applicability of the theory expounded in Ref. 2, we shall consider the following problem: We have a quantum system that is excited by an external field, and is also acted upon by the environment. The starting point in the investigation of such a system is the consideration of the set of Schrödinger operators $\hat{A}_S, \hat{B}_S, \ldots$ for the observable quantities $A(t), B(t), \ldots$ and the equation

$$\frac{\partial\hat{\rho}_1(t)}{\partial t} = \frac{1}{i\hbar}[\hat{H}, \hat{\rho}_1] + \hat{I}(\hat{\rho}_1) \qquad (1)$$

for the density matrix $\hat{\rho}_1(t)$. The subscript 1 indicates that $\hat{\rho}_1(t)$ is the "usual" one-time density matrix (see Sec. 2 below). The matrix $\hat{\rho}_1(t)$ is dimensionless and normalized to unity: $\mathrm{Sp}\,\hat{\rho}_1(t) = 1$. In (1) $\hat{H} = \hat{H}_e + \hat{H}_{\rm int}$, where $\hat{H}_e$ is the proper Hamiltonian of the system, $\hat{H}_{\rm int}$ is the Hamiltonian describing the interaction of the system with the external field, and the operator $\hat{I}(\hat{\rho}_1)$ describes the action of the medium on the systems. Equation (1) is similar to the classical kinetic equation for the distribution function of a gas that is an impurity of low concentration in a denser environment.

It is necessary for us to construct a quantum theory of the fluctuations of the observable quantities in systems described by Eq. (1).

If there is no dissipation, and we must set $\hat{I}(\hat{\rho}_1) = 0$ in (1), then we can construct the quantum-fluctuation theory with the aid of Heisenberg operators of the type

$$\hat{A}_H(t_2t_0) = \exp\left[\frac{i\hat{H}(t_2-t_0)}{\hbar}\right]\hat{A}_S\exp\left[-\frac{i\hat{H}(t_2-t_0)}{\hbar}\right], \qquad (2)$$

where t_0 is the instant of transition from the Schrödinger to the Heisenberg representation. In this case the correlation for the two observable quantities $A(t_2)$ and $B(t_1)$ is given by the well-known formula

$$K[A(t_2)B(t_1)] = \mathrm{Sp}\{{}^1/{}_2\hat{\rho}_1(t_0)[\hat{A}_H(t_2t_0)\hat{B}_H(t_1t_0) + \hat{B}_H(t_1t_0)\hat{A}_H(t_2t_0)]\}. \qquad (3)$$

2. THE DOUBLE-TIME DENSITY MATRIX

Let us derive an expression generalizing (3) to the case when there is dissipation: here we shall assume that the collision operator $\hat{I}$ is a linear function of $\hat{\rho}_1$. In matrix form the collision operator can be written as $I(mnkl)\rho_1(tlk)$. We write the matrix indices as arguments, and sum over the dummy indices. Since Eq. (1) is linear in $\hat{\rho}_1$, its solution can be expressed as a linear function of the initial value of $\hat{\rho}_1(t_0)$:

$$\rho_1(tmn) = W(tmn|t_0m_0n_0)\rho_1(t_0n_0m_0). \qquad (4)$$

The evolution operator $\hat{W}$ satisfies Eq. (1) for the time t and the indices m and n; the time t_0 and the indices m_0 and n_0 are treated in this case as parameters. The initial condition for $\hat{W}$ has the form

$$W(t_0mn|t_0m_0n_0) = \delta_{mn_0}\delta_{nm_0}. \qquad (5)$$

The observable quantity $A(t)$ is equal to

$$A(t) = \mathrm{Sp}(\hat{\rho}_1(t)\hat{A}_S) = W(tmn|t_0m_0n_0)\rho_1(t_0n_0m_0)A_S(nm) = \mathrm{Sp}(\hat{\rho}_1(t_0)\hat{A}_H(tt_0)).$$

We have introduced the Heisenberg operator

$$A_H(tt_0,\ m_0n_0) \equiv W(tmn|t_0m_0n_0)A_S(nm). \qquad (6)$$

When there is no dissipation (i.e., when $\hat{I} = 0$ in (1)), the evolution operator $\hat{W}$ can be expressed in terms of $\hat{H}$ only:

$$W(tmn|t_0m_0n_0) = \exp\left(\frac{-i(t-t_0)\hat{H}}{\hbar}\right)_{mn_0}\exp\left(\frac{i(t-t_0)\hat{H}}{\hbar}\right)_{m_0n}.$$

In this case (6) coincides with (2), and the Heisenberg operators possess the following important property: if $\hat{C}_S = \hat{A}_S\hat{B}_S$, then $\hat{C}_H = \hat{A}_H\hat{B}_H$. In the general case, when allowance is made for the dissipation, the Heisenberg operators (6) no longer possess this property. It is only this circumstance that casts doubt on the possibility of their use in the formula (3).

0038-5646/87/070030-06$04.00

We shall construct a quantum theory of fluctuations for systems with dissipation in a form that is as close as possible to that of the classical theory using the double-time distribution function.[3] Let there be an ensemble consisting of N classical systems, let $\Gamma_i(t)$ be the set of dynamical variables of the system i at the moment t, and let $A(\Gamma_i)$ and $B(\Gamma_i)$ be certain functions of Γ_i. Let us introduce the correlator

$$K[A(t_2)B(t_1)]=\frac{1}{N}\sum_{i=1}^{N} A(\Gamma_i(t_2))B(\Gamma_i(t_1)). \tag{7}$$

There are, besides (7), correlators of a different type with a physical meaning:

$$K[A(t_2)B(t_1)]=\frac{1}{N(N-1)}\sum_{i=1}^{N}\sum_{k\neq i} A(\Gamma_i(t_2))B(\Gamma_k(t_1)), \tag{8}$$

that arise in, for example, the computation of gas-density fluctuations.[4] In this paper we shall consider only the correlators of the type (7) (see Sec. 4 below).

In the quantum theory the correlator corresponding to the classical correlator of the type (7) can be expressed linearly in terms of the operators $\hat{A}_S$ and $\hat{B}_S$, and, consequently, can be written in the form

$$K[A(t_2)B(t_1)]=\rho_2(t_2m_2n_2|t_1m_1n_1)A_S(n_2m_2)B_S(n_1m_1). \tag{9}$$

We shall call the set of quantities $\rho_2(t_2m_2n_2|t_1m_1n_1)$ a double-time density matrix.

Our immediate problem is to show from the first principles of quantum theory that $\hat{\rho}_2$ can be expressed in terms of the evolution operator $\hat{W}$ and the one-time density matrix operator $\hat{\rho}_1$. For this purpose let us consider the closed system consisting of the quantum system in question and the environment (the heat bath). We shall write all the operators and density matrices with double indices of the type $m\alpha$, where the Latin indices pertain to the system in question, while the Greek indices pertain to the heat bath. If the operator $\hat{A}_S$ corresponds to a physical quantity characterizing only the system in question, then it has the form

$$A_S(m\alpha n\beta)=A_S(mn)\delta_{\alpha\beta},$$

where $\delta_{\alpha\beta}$ is the Kronecker symbol. Let us introduce the notation

$$U(tt_0,\, m\alpha n\beta)=\left[\exp\left(\frac{i(t-t_0)\hat{\mathscr{H}}}{\hbar}\right)\right]_{m\alpha n\beta},$$

where $\hat{\mathscr{H}}$ is the total Hamiltonian of the closed system.

According to the first principles of quantum theory, the correlator for two quantities A and B is given by the formula (3), where the Heisenberg operators $\hat{A}_H$ and $\hat{B}_H$ are formed with the aid of the Hamiltonian $\hat{\mathscr{H}}$. If the quantities A and B pertain only to the system under study, then (3) can, without loss of generality, be rewritten in the form (9) with the double-time matrix

$$\begin{aligned}\rho_2(t_2m_2n_2|t_1m_1n_1)={}&{}^1/_2\rho_1(t_0m_0\alpha n_0\beta)[U(t_2t_0,\, n_0\beta n_2\gamma)\\ &\times U(t_0t_2,\, m_2\gamma l\varepsilon)U(t_1t_0,\, l\varepsilon n_1\sigma)U(t_0t_1,\, m_1\sigma m_0\alpha)+U(t_1t_0,\, n_0\beta n_1\sigma)\\ &\times U(t_0t_1,\, m_1\sigma l\varepsilon)U(t_2t_0,\, l\varepsilon n_2\gamma)U(t_0t_2,\, m_2\gamma m_0\alpha)].\end{aligned}$$

Using relations of the type

$$U(t_2t_0,\, n_0\beta n_2\gamma)=U(t_1t_0,\, n_0\beta g\omega)U(t_2t_1,\, g\omega n_2\gamma),$$

we obtain by means of identity transformations the relation

$$\begin{aligned}\rho_2(t_2m_2n_2|t_1m_1n_1)={}&{}^1/_2[\rho_1(t_1m_1\sigma g\omega)U(t_2t_1,\, g\omega n_2\gamma)\\ &\times U(t_1t_2,\, m_2\gamma n_1\sigma)+\rho_1(t_1g\omega n_1\sigma)\\ &\times U(t_2t_1,\, m_1\sigma n_2\gamma)U(t_1t_2,\, m_2\gamma g\omega)].\end{aligned}$$

The one-time density matrix of the system under study is equal to $\rho_1(tmn)=\rho_1(tm\alpha n\alpha)$. In the present investigation we limited ourselves from the very beginning to those conditions under which the system in question can be described not only with the aid of the exact equation for $\rho_1(tm\alpha n\beta)$, which is invariant under time reversal (i.e., under the replacement $t\to -t$), but also with the aid of the approximate equation (1), which contains only $\rho_1(tmn)$ and is, generally speaking, not t invariant, since it describes the dissipation. A detailed derivation of Eq. (1) from the exact equation for $\rho_1(tm\alpha n\beta)$ has been given by, for example, Blum,[5] who uses the approximate equality

$$\rho_1(tm\alpha n\beta)=\rho_1(tmn)\rho_b(\alpha\beta), \tag{10}$$

where $\rho_b(\alpha\beta)$ is the heat bath's constant density matrix, which has the Gibbs form (the latter circumstance will be of no importance to us). Although (10) indicates that the system under study does not interact with the heat bath and, consequently, evolves without dissipation, the approximation (10) can be used at some stages of computations performed with the object of taking account of the dissipation (see Ref. 5).[1)] We shall not discuss the question of the physical meaning and region of applicability of approximations of the type (10), which are used explicitly or implicitly in the derivation of the approximate dissipative equations of the type of the Boltzmann equation or the Bloch equations (1) from the exact equations. To us the important thing is that the approximation (10) has been essentially used in the derivation of (1). Since there is necessarily a region of applicability for (1), and we limited our analysis from the very begining to only this region, we can use (10) in our computations, since we then do not go beyond the scope of those approximations which are always made as soon as we turn to Eq. (1). We have the exact equality

$$\rho_1(tm\alpha n\beta)=U(t_0t,\, m\alpha n_0\omega)\rho_1(t_0n_0\omega m_0\sigma)U(tt_0,\, m_0\sigma n\beta).$$

Hence

$$\rho_1(tmn)=U(t_0t,\, m\alpha n_0\omega)U(tt_0,\, m_0\sigma n\alpha)\sigma_1(t_0n_0\omega m_0\sigma).$$

Using (10) and the last expression for $\rho_1(tmn)$, we obtain the formula (4), with

$$W(tmn|t_0m_0n_0)=U(t_0t,\, m\alpha n_0\omega)U(tt_0,\, m_0\sigma n\alpha)\rho_b(\omega\sigma). \tag{11}$$

Taking account of (10) and (11), we finally find

$$\begin{aligned}\rho_2(t_2m_2n_2|t_1m_1n_1)={}&{}^1/_2[\rho_1(t_1m_1q)W(t_2m_2n_2|t_1qn_1)\\ &+\rho_1(t_1qn_1)W(t_2m_2n_2|t_1m_1q)].\end{aligned}$$

Expressing $\hat{\rho}_1(t_1)$ in terms of $\hat{\rho}_1(t_0)$, and using (9), we can write the correlator in the form

$$\begin{aligned}K[A(t_2)B(t_1)]={}&{}^1/_2A_S(n_2m_2)B_S(n_1m_1)\rho_1(t_0n_0m_0)\\ &\times[W(t_2m_2n_2|t_1qn_1)W(t_1m_1q|t_0m_0n_0)\\ &+W(t_2m_2n_2|t_1m_1q)W(t_1qn_1|t_0m_0n_0)].\end{aligned} \tag{12}$$

If the system under study does not interact at all with the heat bath, i.e., if there is no dissipation, then the evolution operator $\hat{W}$ can be expressed in terms of the Hamiltonian $\hat{H}$

of the system in question. In this case the expression (12) can be rewritten in several equivalent forms, and the relation connecting the times t_2, t_1, and t_0 can be arbitrary. In the presence of dissipation Eq. (1) does not possess time-reversal symmetry. The evolution operator $W(tmn|t_0m_0n_0)$ describes the actual evolution of the physical system in time only when $t \geqslant t_0$. But if we consider the region $t<t_0$ to be admissible, then we can obtain clearly incorrect results, e.g., the result that the occupancy of some level will turn out to be greater than unity (see the formula (16), in which the occupancy $p(t_2|1t_1)>1$ if $t_2<t_1$). Therefore, if, in making allowance for the dissipation, we write the expression for the correlator in the form (12), then we should assume that $t_2 \geqslant t_1 \geqslant t_0$. On the other hand, if, in formulating the problem, we assume that $t_2 \geqslant t_1 \geqslant t_0$, and require that the operators $\hat{W}$ describe the evolution from the smaller to the greater t, then we obtain precisely the expression (12) within the framework of the conclusion drawn above. We assume that the formula (12) solves the problem of the computation of a correlator of the type (7) for a quantum system that interacts with the surrounding medium, and is described by Eq. (1).

It is important that, in deriving (12), we used only the approximation (10), which has already been used by Blum[5] to derive (1), and does not infringe upon the first principles of quantum theory. Therefore, it should be assumed that the formula (12) is in accord with these principles, and that its region of applicability coincides with that of Eq. (1). Any derivation of the equations taking account of the dissipation from the exact equations, which are invariant under time reversal, contains assumptions, the meaning and region of applicability of which are not quite clear. Consequently, all the equations that take account of the dissipation, in particular, Eqs. (1) and (12), should, strictly speaking, be regarded as a hypothesis, the validity of which can, in the final analysis, be verified only through comparison with experiment. Below, in Sec. 4, we indicate the observable consequences of (12).

The procedure proposed by us here for the computation of the correlators can also be used in the case when the operators $\hat{H}$ and (or) $\hat{I}$ in Eq. (1) are time dependent. In this case only certain formulas, e.g., (2) and (3), which, for simplicity, are given here for the case when $\hat{I}=0$ and $\hat{H}$ does not depend on the time, change.

3. HEAT-INDUCED FLUCTUATIONS

Let us introduce the operator describing the heating of the medium. For this purpose let us compute the time derivative of the energy E of the system. We have

$$\frac{dE}{dt}=\mathrm{Sp}\left(\frac{d\hat{\rho}_1(t)}{dt}\hat{H}_e\right)=\mathrm{Sp}(\hat{\rho}_1(t)\hat{P}_S)-\mathrm{Sp}(\hat{\rho}_1(t)\hat{Q}_S),$$

where the operator $\hat{P}_S=(1/i\hbar)[\hat{H}_e,\hat{H}_{\mathrm{int}}]$ describes the variation of the energy of the system under the action of the external field, and the operator

$$Q_S(mn)=-H_e(qp)I(pqmn)$$

describes the variation of the energy of the system under the action of the medium. We assume that the energy lost by the system as a result of the interaction with the medium is acquired by the medium. Since the energy lost per unit time is equal to $Q(t)=\mathrm{Sp}(\hat{\rho}_1(t)\hat{Q}_S)$, the Schrödinger operator $\hat{Q}_S$ should be called the medium-heating operator.

Below in specific computations we shall consider a two-level system that is excited by a periodic electric field $\vec{\mathscr{E}}$. In this case in the matrix representation in the basis of stationary states

$$\hat{H}_e=\begin{pmatrix}E_0 & 0\\ 0 & E_1\end{pmatrix},\quad \hat{H}_{int}=-\begin{pmatrix}0 & \mathbf{d}_{01}\vec{\mathscr{E}}\\ \mathbf{d}_{10}\vec{\mathscr{E}} & 0\end{pmatrix},$$

where E_0 and E_1 are the energies of the ground and excited states, respectively, and $\mathbf{d}_{01}=\langle 1|\mathbf{d}|0\rangle$ is the dipole moment of the transition between these states. Let us assume that $I(0011)=-I(1111)=1/\tau$, $I(0101)=I(1010)=-1/T$, and that the remaining components of $\hat{I}$ are equal to zero. It is easy to see that τ is the system's energy dissipation time and T is the phase relaxation time. In this case, for a two-level system, the heating operator has the form

$$\hat{Q}_S=\begin{pmatrix}0 & 0\\ 0 & \dfrac{E_1-E_0}{\tau}\end{pmatrix}. \tag{13}$$

Let the electric field be linearly polarized, i.e., let $\vec{\mathscr{E}}(t)=\mathbf{e}_l\mathscr{E}_l(t)\cos\omega_l t$, with $|\omega_l-\omega_e|\ll\omega_e$, where $\omega_e=(E_1-E_0)/\hbar$ is the eigenfrequency of the system, $\mathbf{e}_l$ is the polarization vector, and the amplitude $\mathscr{E}_l(t)$ is, generally speaking, time dependent. Let the phase relaxation occur so rapidly that $T\mathscr{E}_l d_{01}/\hbar \ll 1$. Then, as is well known, the excitation of the system occurs in an incoherent manner, and Eq. (1) reduces to the form

$$\frac{d}{dt}\begin{bmatrix}1-p & \rho_{01}\\ \rho_{10} & p\end{bmatrix}=\begin{bmatrix}-w(1-2p)+\nu p & -\dfrac{\rho_{01}}{T}\\ -\dfrac{\rho_{10}}{T} & w(1-2p)-\nu p\end{bmatrix}. \tag{14}$$

We have introduced the notation $\nu\equiv\tau^{-1}$ for the rate of dissipation and $p\equiv\rho_{11}$ for the occupancy of the top level; the rate w of excitation is equal to $w=F\sigma$, where

$$\sigma=2\pi\frac{\omega_l}{\hbar c}\frac{d_{01}{}^2T}{1+T^2(\omega_l-\omega_e)^2}$$

can be interpreted as the cross section for photon absorption and $F=c\mathscr{E}_l^2/(8\pi\hbar\omega_l)$ as the photon flux. Notice that $w\propto\mathscr{E}_l^2(t)$; in a field with a variable amplitude $\mathscr{E}_l(t)$ the excitation rate $w(t)$ is time dependent.

Let $p(t|t_0p_0)$ be the occupancy of the top level at the moment t under the condition that this occupancy was equal to p_0 at the moment t_0. If the rate excitation is time independent, then from (14) we immediately obtain $(t>t_0)$

$$p(t|t_0p_0)=\frac{w}{2w+\nu}+\exp[-(2w+\nu)(t-t_0)]\left(p_0-\frac{w}{2w+\nu}\right). \tag{15}$$

When the difference $t-t_0$ is much greater than the characteristic transient period $(2w+\nu)^{-1}$, the occupancy p does not depend on the initial value p_0, and is equal to the steady-state value $p_s=w/(2w+\nu)$. The corresponding steady-state value Q_s of the heating rate is equal to

$$Q_s=\frac{\hbar\omega_e}{\tau}\frac{w}{2w+\nu}.$$

Let us find the evolution operator from the Eq. (14)

and the initial condition (5). Then let us find the heating correlator from the formula (12). Let the time $t_0 \to -\infty$. Then the correlator does not depend on the initial value $\hat{\rho}_1(t_0)$ of the density matrix, and it equal to

$$K[Q(t_2)Q(t_1)] = \left(\frac{\hbar\omega_e}{\tau}\right)^2 p(t_2|t_1 1)p_s(t_1), \tag{16}$$

where $p_s(t_1) = w(t_1)/(2w(t_1)+\nu)$ is the steady-state occupancy of the top level at the moment t_1 and $w(t_1)$ is the rate of excitation at the moment t_1.

Let the medium be irradiated by incident light pulses of duration t_l (in Ref. 1 $t_l = 2\times 10^{-8}$ sec). We shall assume that the characteristic variation time for $w(t)$ is also of the order of t_l; this time is much longer than the characteristic transient period $(2w+\nu)^{-1}$. In this case we can, with an accuracy sufficient for our purposes, replace (16) by

$$K[t_2t_1] = \left(\frac{\hbar\omega_e}{\tau}\right)^2 \left\{\frac{w(t_2)w(t_1)}{(2w(t_2)+\nu)(2w(t_1)+\nu)} + \frac{w(t)}{2w(t)+\nu}\left(1-\frac{w(t)}{2w(t)+\nu}\right)\exp[-(2w(t)+\nu)(t_2-t_1)]\right\}, \tag{17}$$

where t is some time close to t_1 and t_2. The exact definition of t is not necessary, since, as the difference $t_2 - t_1$ increases, the term containing the exponential function in (17) decreases rapidly.

Let us discuss the result obtained, assuming, for simplicity, that w does not depend on the time. In this case $K[t_2t_1] = K[t_{21}]$, where $t_{21} = t_2 - t_1$. When $t_{21} = 0$, we have $K[0] = \mathrm{Sp}(\hat{\rho}_1\hat{Q}_s^2)$, as it should be. Notice that $K[0] = Q_s^2/p_s$. For $t_{21}\to\infty$, we have $K[\infty] = Q_s^2$, as it should be. Thus, $K[\infty] = K[0]p_s$. For low incident-light intensities, when the excitation rate w is much lower than the dissipation rate ν, the steady-state occupancy of the top level $p_s \ll 1$. In this case $K[\infty] \ll K[0]$, which indicates strong fluctuations in the heating rate $Q(t)$. The realization plot for the stochastic process $Q(t)$ consists of peaks of temporal width of the order or τ, which are randomly disposed along the time axis, with temporal separations of the order of w^{-1}. The maximum value of $Q(t)$ in a peak is, in order of magnitude, equal to $\hbar\omega_e/\tau$; between the peaks the quantity $Q(t)$ is negligibly small. Each peak can be interpreted as the result of the absorption of one photon and the subsequent dissipation of its energy. But such an interpretation is not quite correct, since in our model the electric field of the incident light was described classically. The strong heating fluctuations in the case of low incident-light intensity are a consequence of the quantum nature of the two-level system, which has an essentially discrete energy spectrum and for which a classical description is, in principle, impossible.

At high incident–light intensities we have $w \gg \nu$ and $p_s = \frac{1}{2}$. In this case $K[\infty] = \frac{1}{2}K[0]$. From this it follows that the heating fluctuations are moderate. The plot of $Q(t)$ is modulated to a depth of the order of the mean value of $Q(t)$. But the characteristic modulation time $(2w+\nu)^{-1} \approx (2w)^{-1}$ is much shorter than the characteristic temporal width τ of the peaks that make up the plot of $Q(t)$ in the opposite limiting case. Thus, as the incident-light intensity increases, the fluctuations in the heating of the medium by the two-level impurity particle become less deep, but more rapid.

In conclusion of this section, let us note that, according to the present investigation, at low intensities, when $w \ll \nu$ and the occupancy of the upper level is low, the heating fluctuations occur as described in Ref. 2.

4. INTENSITY AND FREQUENCY SPECTRUM OF THE NTIS

Let us compute the total intensity of the nonlinear thermal-impurity scattering (NTIS) observed by Belyaev *et al.*[1] let us also compute this intensity's frequency distribution, which will be fixed by the photographic plate or the photon counter after the passage of the incident light pulse. The starting point is the Maxwell equation

$$\frac{\varepsilon_0\,\partial^2\vec{\mathscr{E}}_1}{\partial t^2} - c^2\Delta\vec{\mathscr{E}}_1 = -\frac{\partial^2\varepsilon_1(t\mathbf{r})\vec{\mathscr{E}}_l}{\partial t^2}, \tag{18}$$

where $\vec{\mathscr{E}}_l$ and $\vec{\mathscr{E}}_1$ are the electric field intensities of the incident and scattered light and ε_0 and $\varepsilon_1(t\mathbf{r})$ are respectively the constant and fluctuating parts of the permittivity. Equation (18) is linear in ε_1 and $\vec{\mathscr{E}}_1$. Let us, assuming that the fluctuations occur at constant pressure p of the medium, write $\varepsilon_1 = (\partial\varepsilon/\partial T)_p T_1(t\mathbf{r})$, where the fluctuating part $T_1(t\mathbf{r})$ of the temperature is equal to

$$\begin{aligned} T_1(t\mathbf{r}) &= \frac{1}{nc_p}\int d^3r_1\int_0^t dt_1 G_T(\mathbf{r}-\mathbf{r}_1, t-t_1) \\ &\quad\times\sum_i \delta(\mathbf{r}-\mathbf{r}_i(t_1))Q(\Gamma_i(t)) \\ &= \frac{1}{nc_p}\int_0^t dt_1\sum_i G_T(\mathbf{r}-\mathbf{r}_i(t_1), t-t_1)Q(\Gamma_i(t_1)). \end{aligned} \tag{19}$$

Here n is the concentration of the particles of the medium, c_p is the dimensionless constant-pressure specific heat per particle, $Q(\Gamma_i(t))$ is the energy transferred in unit time to the medium from the point particle located at the point $\mathbf{r}_i(t)$ (for convenience of notation, we measure the temperature in energy units, and for the present use the classical theory),

$$G_T(\mathbf{r}, t) = \frac{1}{[2(\pi\chi t)^{1/2}]^3}\exp\left[-\frac{r^2}{4\chi t}\right]$$

is the Green function for the thermal-conductivity equation, and χ is the coefficient of thermal conductivity of the medium. The illumination of the impurity-containing medium began at the moment $t = 0$, and ended at the moment t_l; for each particle i the heating rate $Q(\Gamma_i(t))$ is nonzero only in this time interval.

Next, we shall solve the standard problem of light scattering by the permittivity fluctuations. According to (18), the source of the scattered field $\vec{\mathscr{E}}_1$ is the field of the incident light $\vec{\mathscr{E}}_l$ and the fluctuating part ε_1 of the permittivity on the right-hand side of (18). Using (18) and (19), we find through standard computations carried out in the spirit of the Hamiltonian method[6] the field $\mathscr{E}_1(t,\mathbf{r})$, which can be linearly expressed in terms of the right-hand side of (18). The intensity I of the scattered light is proportional to $\mathscr{E}_1^2$. From this it is clear that the expression for I contains the sum

$$\sum_i\sum_k Q(\Gamma_i(t_1))Q(\Gamma_k(t_2)),$$

which splits up into two sums of the type (7) and (8). We can, by carrying out all the computations, verify that the term containing the sum of the type (8) vanishes if the positions of the impurity particles in space are not correlated. The remaining term gives the expression

$$I(\omega, \mathbf{n}, \sigma) = \frac{N\varepsilon_0^{1/2}|\mathbf{e}_{\mathbf{k}\sigma}{}^*\mathbf{e}_l|^2}{16(2\pi)^4 c^3 c_p{}^2 n^2}\left(\frac{\partial\varepsilon}{\partial T}\right)^2 J. \tag{20}$$

Here $dE = I(\omega,\mathbf{n},\sigma)d\omega d^2n$ is the scattered-light energy that was emitted over the entire pulse duration t_l into the solid angle d^2n in the frequency range $d\omega$ in the form of electromagnetic waves with polarization vector $\mathbf{e}_{\mathbf{k}\sigma}$, where $\sigma = 1$, 2; $\mathbf{k}$ is the wave vector of the scattered light; $\mathbf{n} = \mathbf{k}/k$ is the unit vector in the scattering direction; N is the number of impurity particles in the illuminated volume;

$$J=\int_0^{t_l} dt_1 \int_0^{t_l} dt_2 \mathscr{E}_l(t_1)\mathscr{E}_l(t_2) \int_0^{t_1} dt_3 \int_0^{t_2} dt_4 \int d^3x \int d^3y$$

$$\times G_T(\mathbf{x}, t_1-t_3)\, G_T(\mathbf{y}, t_2-t_4) \cos[\varkappa(t_2-t_1) - \mathbf{q}(\mathbf{x}-\mathbf{y})] K_s[t_4t_3], \tag{21}$$

with $\varkappa = \omega - \omega_l$ and $\mathbf{q} = \mathbf{k} - \mathbf{k}_l$; let us recall that ω_l and $\mathbf{k}_l$ are respectively the frequency and wave vector of the incident light. The symmetrized heating correlator is equal to

$$K_s[t_4t_3] = \theta(t_4-t_3)K[t_4t_3] + \theta(t_3-t_4)K[t_3t_4], \tag{22}$$

where $\theta(t)$ is the Heavyside step function and $K[t_4t_3]$ is defined according to (7). The quantum nature of the motion involving the internal degrees of freedom of the impurity particle does not change anything in the formulas (20)–(22), but as the heating correlator in (22) we should, within the framework of our model, substitute the expression (17).

The expressions (17) and (20)–(22) completely determine the intensity $I(\omega,\mathbf{n},\sigma)$ of the NTIS. The facor J contains the dependence of $I(\omega,\mathbf{n},\sigma)$ on all the important parameters of the problem, except the polarization [see (20) and (21)].

Let us consider the NTIS in the case when the scattering angle θ between the vectors $\mathbf{k}$ and $\mathbf{k}_l$ is so small that we can set $\mathbf{q} = \mathbf{k} - \mathbf{k}_l \approx 0$ in (21), and immediately carry out the $d^3\mathbf{x}$ and $d^3\mathbf{y}$ integrations, using the equality $\int d^3x G_T(\mathbf{x},t) = 1$. Under the conditions of the experement reported in Ref. 1, this requires that $\theta < 20$ or 25°. Concerning the scattering at large angles, we only note that the NTIS intensity should decrease with increasing θ because of the presence of a rapidly oscillating factor in (21).

For the small-angle scattering the subsequent computations are carried out with the aid of standard mathematical methods. We shall give only the results here. We shall find the dependence of the NTIS intensity on the characteristic incident-light intensity I_l and pulse duration t_l by carrying out in (21) integration over all the frequency detunings $\varkappa$. The factor $J(\varkappa,\theta)$ does not depend on the scattering angle θ if this angle is small. Let us introduce the notation $\Phi = \int d\varkappa J(\varkappa)$. Usually, the dissipation rate $\nu \approx 10^{12}$ sec^{-1}, which is much greater than $t_l^{-1} = 5\times 10^7$ sec^{-1}; therefore, the expression (21) should be analyzed in the three limiting cases for the dependence on the characteristic value of the excitation rate $\bar{w}$: if

$$\begin{aligned}&\bar{w} \ll t_l^{-1} \ll \nu \text{ (weak excitation), then } \Phi \propto I_l^2 t_l^2,\\ &t_l^{-1} \ll \bar{w} \ll \nu \text{ (moderate excitation), then } \Phi \propto I_l^3 t_l^3,\\ &t_l^{-1} \ll \nu \ll \bar{w} \text{ (strong excitation), then } \Phi \propto I_l t_l^3.\end{aligned} \tag{23}$$

Belyaev *et al.*[1] observed the dependence $\Phi \propto I_l^3$, which corresponds to the case of moderate excitation.

Let us find the frequency spectrum of the NTIS, i.e., the dependence of $J(\varkappa)$ on the frequency detuning $\varkappa$. We shall, depending on the magnitude of the detuning, call the sections of the NTIS spectral line as follows: $|\varkappa| \lesssim t_l^{-1}$ the line center, $t_l^{-1} \ll |\varkappa| \lesssim 2\bar{w} + \nu$ the line slope, and $2\bar{w} + \nu \lesssim |\varkappa|$ the line wings.

Let us perform the dt_1 and dt_2 integrations in (21) by parts, and then make the substitutions $t_1 \to t - z/2$ and $t_2 \to t + z/2$. We obtain

$$\begin{aligned}J = \frac{1}{\varkappa^2}\int_0^{t_l} dt \int_{-(t_l-|2t-t_l|)}^{t_l-|2t-t_l|} dz \cos\varkappa z\Big\{&\mathscr{E}_l\left(t+\frac{z}{2}\right)\mathscr{E}_l\left(t-\frac{z}{2}\right)\\ &\times K_s\left[t+\frac{z}{2}, t-\frac{z}{2}\right]\\ &+\mathscr{E}_l'\left(t+\frac{z}{2}\right)\mathscr{E}_l\left(t-\frac{z}{2}\right)\int_0^{t-z/2} dt_3 K_s\left[t+\frac{z}{2}, t_3\right]\\ &+\mathscr{E}_l\left(t+\frac{z}{2}\right)\mathscr{E}_l'\left(t-\frac{z}{2}\right)\int_0^{t+z/2} dt_3 K_s\left[t-\frac{z}{2}, t_3\right]\\ &+\mathscr{E}_l'\left(t+\frac{z}{2}\right)\mathscr{E}_l'\left(t-\frac{z}{2}\right)\int_0^{t+z/2} dt_3 \int_0^{t-z/2} dt_4 K_s[t_4, t_3]\Big\},\end{aligned} \tag{24}$$

where the prime denotes differentiation with respect to the argument. The frequency spectrum of the NTIS at the line center varies slowly because of the absence of large analytic parameters in (21) and (24). On the slope and at the wings of the line, i.e., in the region where $|\varkappa| t_l \gg 1$, the frequency spectrum of the NTIS is determined by the distant Fourier components of the expression in the curly brackets in (24). The correlator $K_s[t + z/2, t - z/2]$ and, along with it, the first term in the curly brackets as functions of z, have first-derivative discontinuities at the point $z = 0$. The other terms are smoother functions of z, and they can be discarded in the evaluation of the dz integral. Next, let us use (22) and (17), with account taken in (17) of only the term containing the exponential function and furnishing the first-derivative discontinuity. Setting $\mathscr{E}_l(t + z/2)\mathscr{E}_l(t - z/2) \approx \mathscr{E}_l^2(t)$, and extending the limits of the dz integration to infinity, we obtain

$$J(\varkappa) = \frac{2}{\varkappa^2}\left(\frac{\hbar\omega_e}{\tau}\right)^2$$

$$\times \int_0^{t_l} dt \mathscr{E}_l{}^2(t) \frac{w(t)(w(t)+\nu)}{(2w(t)+\nu)[(2w(t)+\nu)^2+\varkappa^2]}. \tag{25}$$

According to (25), on the line slope we have, depending on the magnitude of the excitation, the following estimates for J: In the case of weak and moderate excitations

$$J \propto I_l^2 t_l/\varkappa^2, \tag{26a}$$

while in the case of strong excitation

$$J \propto t_l/\varkappa^2. \tag{26b}$$

At the wings of the line we have for any excitation

$$J \propto I_l^2 t_l / \varkappa^4. \qquad (27)$$

Notice that the dependence of J on t_l and I_l on the slope and at the wings of the line differs from the corresponding dependence for the total line intensity.

As can be seen from the foregoing, there exists the critical value $\varkappa_c = 2\bar{w} + \nu$, where $\bar{w}$ is the characteristic value of the excitation rate. When the frequency detuning $\varkappa$ passes through this value, the dependence $J(\varkappa)$ changes (see (26) and (27)). Since $\varkappa_c \propto I_l$, the NTIS spectral line width increases linearly with the incident-light intensity I_l.

The assertions (23), (26a), (26b), and (27) are entirely accessible to experimental verification.

5. CONCLUSION

In one article it is impossible not alone to investigate, but even to indicate, all the relations between the above-developed theory of temporal fluctuations in dissipative quantum systems and the related areas of physics. We only note that the various aspects of the theory of fluctuations have been investigated in a large number of papers (see, for example, Refs. 7–14), but not one of these papers considers in its explicit form the problem of interest to us, which is formulated and solved in Secs. 1 and 2 of the present paper.

We have considered the physical situation in which the heating of the transparent medium by the radiation proceeds via microscopic impurity particles (atoms or molecules), which play the role of intermediaries between the radiation and the medium. The heating fluctuations are not connected with the quantum nature of the radiation, but are due to the discreteness of the energy spectrum of the impurity particles. If we model the impurity particle by a linear oscillator, and assume that the radiation excites the oscillator to high levels, where the discreteness is unimportant, then the heating fluctuations vanish[15].

In the other physical situation in which the heating proceeds via impurity grains or other absorbing macroscopic-type centers, temporal heating fluctuations can also occur, but they occur for other reasons (see, for example, Ref. 16 and the literature indicated therein).

The author thanks V. L. Ginzburg and A. B. Klimov for a discussion. He also thanks all the participants of the seminars conducted by E. S. Fradkin, E. E. Nikitin, S. M. Rytov and V. I. Tatarskiĭ, D. A. Kirzhnits, M. L. Ter-Mikaelyan, A. M. Bonch-Bruevich, where this paper was discussed.

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1) This type of inconsistency is often encountered in physical computations. For example, the electron-energy losses that occur during synchrotron radiation are often computed under the assumption that the electron moves along a circle, and not along a convergent spiral. Although motion along a circle implies that the electron does not lose energy, the circular-motion approximation can be used if the energy losses per revolution are small, and the synchrotron-radiation intensity is what is actually computed.

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Translated by A. K. Agyei