Correlation effects in cooperative Raman scattering of light

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A new approach to the quantum theory of spatiotemporal evolution of cooperative Raman scattering of light is proposed, with space correlations taken into account. The role of these correlations in scattering under various scattering conditions is elucidated. The spatial distributions of the scattered-field intensities and of the correlation functions are investigated.

It is common knowledge that a distinctive feature of cooperative phenomena in many-atom systems is the presence of two stages. The first corresponds to the development of spontaneous polarization, when the population of the initial level is high and the number of emitted photons is small. During the second stage, when the number of emitted photons approaches the value of the initial population, propagation sets in of radiation that interacts resonantly with the medium. The first stage requires a consistent quantum description,[1–5] while the second can be described by a semiclassical approach.[6] The emitted-pulse parameter fluctuations due to the system dynamics during the first stage are quite appreciable. The reason is that the correlation evolving during the first stage covers macroscopic regions of the sample. It follows from the foregoing that an adequate description of cooperative scattering during all stages of its development requires a quantum approach with allowance for propagation effects.

We develop here for cooperative phenomena a consistent theory based on the use of two-point correlation functions. The obtained system of equations permits an investigation of both the evolution of the correlation function in the initial stage and the spatial evolution of the field in the pulse-onset stage.

The system investigated consists of an electromagnetic field and an atomic subsystem. We describe the field by a vector-potential operator $\mathbf{A}$ and its conjugate general momentum

$$\mathbf{B}=\frac{1}{4\pi c^2}\frac{\partial\mathbf{A}}{\partial t}.$$

They satisfy the following commutation relations[7]

$$[A_\alpha(\mathbf{r},t),A_\beta(\mathbf{r}',t)]=[B_\alpha(\mathbf{r},t),B_\beta(\mathbf{r}',t)]=0,$$
$$[A_\alpha(\mathbf{r},t),B_\beta(\mathbf{r}',t)]=i\hbar\delta_{\alpha\beta}\delta(\mathbf{r}-\mathbf{r}'). \tag{1}$$

The subsystem of N two-level atoms (transition frequency ω_0) is described by the spin operators σ_+, σ_-, and σ_3, where

$$\sigma_\pm=(\sigma_1\pm i\sigma_2)/2,$$
$$[\sigma_+,\sigma_-]=\sigma_3,\ [\sigma_\pm,\sigma_3]=\mp2\sigma_\pm.$$

The Hamiltonian of such a system is of the form

$$H=H_f+H_a+H_{\rm int}, \tag{2}$$

$$H_f=\int\left[2\pi c^2\mathbf{B}^2+\frac{1}{8\pi}(\mathrm{rot}\,\mathbf{A})^2\right]dV$$

is the electromagnetic-field Hamiltonian,

$$H_a=\sum_{i=1}^{N}\frac{\hbar\omega_0}{2}\sigma_3^{\,i}$$

is the atomic-system Hamiltonian, and

$$H_{\rm int}=-\frac{1}{c}\int\mathbf{j}(\mathbf{r},t)\mathbf{A}(\mathbf{r},t)\,dV$$

is the interaction Hamiltonian. Here

$$\mathbf{j}(\mathbf{r},t)=\sum_{i=1}^{N}[\langle+|\mathbf{j}(\mathbf{r}_i,t)|-\rangle\sigma_+^{\,i}+\langle-|\mathbf{j}(\mathbf{r}_i,t)|+\rangle\sigma_-^{\,i}]\mathbf{E}_p(\mathbf{r}_i,t)$$

is the current density in the presence of pumping (frequency ω_p, wave vector $\mathbf{k}_p$),

$$\mathbf{E}_p=\mathbf{A}_{p0}\exp[i(\mathbf{k}_p\mathbf{r}-\omega_pt)]+\text{h.c.}$$

is the c-number pump field, and $|+\rangle$, and $|-\rangle$ are the wave functions of the excited and ground states of the atoms, respectively.

We represent $\mathbf{A}(\mathbf{r},t)$ as a sum of vector components of a Stokes field (frequency ω_s, wave vector $\mathbf{k}_s$ and an anti-Stokes field (frequency ω_a, wave vector $\mathbf{k}_a$, and separate in them the rapidly oscillating factors

$$\mathbf{A}(\mathbf{r},t)=\mathbf{A}_s^{+}(\mathbf{r},t)\exp(i\omega_st)+\text{h.c.}+\mathbf{A}_a^{+}(\mathbf{r},t)\exp(i\omega_at)+\text{h.c.},$$

where

$$|\partial\langle\mathbf{A}^{\pm}_{s(a)}\rangle/\partial t|\ll\omega_{s(a)}\langle\mathbf{A}^{\pm}_{s(a)}\rangle.$$

Then

$$\mathbf{B}(\mathbf{r},t)=\frac{i\omega_s}{4\pi c^2}[\mathbf{A}_s^{+}(\mathbf{r},t)\exp(i\omega_st)-\text{h.c.}]+\frac{i\omega_a}{4\pi c^2}[\mathbf{A}_a^{+}(\mathbf{r},t)\exp(i\omega_at)-\text{h.c.}].$$

Carrying out a canonical transformation, as in Ref. 7, and changing from the variables $\mathbf{A}$ and $\mathbf{B}$ to the variables $\mathbf{A}^+$ and $\mathbf{B}^-=-(i\omega/2\pi c^2)\mathbf{A}^-$, we reduce the Hamiltonian (2) ultimately to the form

$$H=H_f^{\,s}+H_f^{\,a}+H_a+H_{\rm int}, \tag{3}$$

where

$$H_f^{\,s(a)}=\frac{1}{8\pi}\int\left[\frac{\omega^2_{s(a)}}{c^2}(\mathbf{A}^{+}_{s(a)}\mathbf{A}^{-}_{s(a)}+\text{h.c.})+(\mathrm{rot}\,\mathbf{A}^{+}_{s(a)}\,\mathrm{rot}\,\mathbf{A}^{-}_{s(a)}+\text{h.c.})\right]dV,$$

$$H_{int}=-\frac{1}{c}\sum_{j=1}\Big\{[g^*\sigma_+{}^jA_a{}^-(\mathbf{r}_j,t)$$

$$\times\exp(-i\mathbf{k}_p\mathbf{r}_j+i(\omega_p-\omega_a)t)+\text{h.c.}]$$

$$+[g\sigma_+{}^jA_s{}^+(\mathbf{r}_j,t)\exp(i\mathbf{k}_p\mathbf{r}_j+i(\omega_s-\omega_p)t)+\text{h.c.}]\Big\},$$

$$g=\langle+|\mathbf{j}^+(\mathbf{r})|-\rangle A_{p0}{}^-.$$

The commutation relations for $\mathbf{A}^+_{s(a)}$ and $\mathbf{A}^-_{s(a)}$ are obtained from (1)

$$[\mathbf{A}^+_{s(a)\alpha}(\mathbf{r},t),\mathbf{A}^+_{s(a)\beta}(\mathbf{r}',t)]=[\mathbf{A}^-_{s(a)\alpha}(\mathbf{r},t),\mathbf{A}_{s(a)\beta}(\mathbf{r}',t)]=0,$$

$$[\mathbf{A}^+_{s(a)\alpha}(\mathbf{r},t),\mathbf{A}^-_{s(a)\beta}(\mathbf{r}',t)]=\frac{2\pi\hbar c^2}{\omega_{s(a)}}\delta_{\alpha\beta}\delta(\mathbf{r}-\mathbf{r}'). \quad (4)$$

From the Heisenberg equation for the operators

$$i\hbar\frac{\partial f}{\partial t}=[f,H]$$

with the Hamiltonian (3) and the commutation relations (4) we can obtain the following equations:

$$\frac{\partial\mathbf{A}^+_{s(a)}}{\partial t}=-\frac{i\omega_{s(a)}}{2}\mathbf{A}^+_{s(a)}+\frac{ic^2}{2\omega_{s(a)}}\text{rot rot}\,\mathbf{A}^+_{s(a)}$$

$$-\frac{2i\pi c}{\omega_{s(a)}}\sum_{j=1}^{N}g^*(g)\sigma_{-(+)}^{\ j}\exp[-i\mathbf{k}_p\mathbf{r}_j+i(\omega_p-\omega_{s(a)})t]\delta(\mathbf{r}-\mathbf{r}_j);$$

$$\frac{\partial\sigma_+{}^j}{\partial t}=i\omega_0\sigma_+{}^j+\frac{i}{\hbar c}g^*\sigma_3{}^j\{A_a{}^+\exp[i\mathbf{k}_p\mathbf{r}_j-i(\omega_p-\omega_a)t]$$

$$+A_s{}^-\exp[-i\mathbf{k}_p\mathbf{r}_j+i(\omega_p-\omega_s)t]\}; \quad (5)$$

$$\frac{\partial\sigma_3{}^j}{\partial t}=\frac{2i}{\hbar c}\{(g\sigma_+{}^jA_a{}^-\exp[-i\mathbf{k}_p\mathbf{r}_j+i(\omega_p-\omega_a)t]-\text{h.c.})$$

$$+(g\sigma_+{}^jA_s{}^+\exp[i\mathbf{k}_p\mathbf{r}_j+i(\omega_s-\omega_p)t]-\text{h.c.})\}.$$

In experiments aimed at observing cooperative Raman scattering (CRS) the sample is usually a thin elongated cylinder. It is useful therefore to seek the solution in the form

$$\mathbf{A}^\pm_{s(a)}(r,t)=\mathbf{A}^\pm_{s(a)0}(r,t)\exp(\mp ik_{s(a)}x), \quad (6)$$

where $k_{s(a)}=(\mathbf{k}_{s(a)})_x$, the x axis is directed along the sample axis, and the $\mathbf{A}^\pm_{s(a)0}$ satisfy the conditions

$$\left|\frac{\partial\langle\mathbf{A}^\pm_{s(a)0}\rangle}{\partial t}\right|\ll k_{s(a)}\langle\mathbf{A}^\pm_{s(a)0}\rangle. \quad (7)$$

Using (6) and (7) we can obtain from (5) a system of equations that take, after averaging over a volume V_1 with dimensions much larger than the scattering wavelength and smaller than the characteristic length $\mathbf{A}^\pm_{s(a)0}$ of amplitude variation, the form

$$\frac{\partial n_{s(a)}(x,x')}{\partial t}+c\left(\frac{\partial}{\partial x}+\frac{\partial}{\partial x'}\right)n_{s(a)}(x,x')$$

$$=\frac{1}{2}[F_{s(a)}(x,x')+F_{s(a)}(x',x)],$$

$$\frac{\partial F_s(x,x')}{\partial t}+c\frac{\partial}{\partial x}F_s(x,x')$$

$$=\frac{1}{T_0^2}\{-2R_3(x')n_s(x,x')+S(x,x')$$

$$+[S_0(x)-S(x,x')]\delta(x-x')-\zeta(x)R_3(x')Q(x,x')\},$$

$$\frac{\partial F_a(x,x')}{\partial t}+c\frac{\partial}{\partial x}F_a(x,x')=\frac{1}{T_0^2}\{2R_3(x')n_a(x,x')$$

$$+A(x,x')+[A_0(x)-A(x,x')]\delta(x-x')$$

$$+\zeta(x)R_3(x')Q(x,x')\},$$

$$\frac{\partial S(x,x')}{\partial t}+\frac{S(x,x')}{T_2}=-R_3(x)F_s(x,x')-R_3(x')F_s(x',x)$$

$$+\zeta(x-x')[R_3(x)F_a(x,x')+R_3(x')F_a(x',x)], \quad (8)$$

$$\frac{\partial A(x,x')}{\partial t}+\frac{A(x,x')}{T_2}=R_3(x)F_a(x,x')+R_3(x')F_a(x',x)$$

$$-\zeta(x-x')[R_3(x)F_s(x,x')+R_3(x')F_s(x',x)],$$

$$\frac{\partial Q(x,x')}{\partial t}+c\left(\frac{\partial}{\partial x}+\frac{\partial}{\partial x'}\right)Q(x,x')$$

$$=-\zeta(x')F_s(x,x')-\zeta(x)F_a(x',x),$$

$$\frac{\partial R_3(x)}{\partial t}+\frac{1}{T_1}R_3(x)=F_s(x,x)-F_a(x,x),$$

where

$$n_{s(a)}(x,x')=\frac{\omega_{s(a)}}{4\pi\hbar c^2}\frac{1}{N_1^2}\sum_{j\in V_1(x)}\sum_{k\in V_1(x')}[\mathbf{A}^+_{s(a)0}(x_j)\mathbf{A}^-_{s(a)0}(x_k)+\text{h.c.}]$$

is the density of the average number of scattering photons, normalized to the atom-number density $N_V=N/V$ (V is the sample volume), $N_1=N_VV_i$,

$$F_s(x,x')=-\frac{i}{\hbar cN_1^2}\sum_{j\in V_1(x)}\sum_{k\in V_1(x')}\{g^*\sigma_-{}^kA_{s0}{}^-(x_j)\exp[i(\omega_p-\omega_s)t+i(k_s-k_p)x_k]-\text{h.c.}\},$$

$$F_a(x,x')=-\frac{i}{\hbar cN_1^2}\sum_{j\in V_1(x)}\sum_{k\in V_1(x')}\{g\sigma_+{}^kA_{a0}{}^-(x_k)\exp[i(\omega_p-\omega_a)t+i(k_a-k_p)x_k]-\text{h.c.}\}.$$

The quantities

$$S(x,x')=\frac{1}{2N_1^2}\sum_{j\in V_1(x)}\sum_{\substack{k\in V_1(x')\\k\neq j}}\{\sigma_-{}^j\sigma_+{}^k\exp[i(k_p-k_s)(x_k-x_j)]+\text{h.c.}\},$$

$$A(x,x')=\frac{1}{2N_1^2}\sum_{j\in V_1(x)}\sum_{\substack{k\in V_1(x')\\k\neq j}}\{\sigma_+{}^j\sigma_-{}^k\exp[i(k_p-k_a)(x_k-x_j)]+\text{h.c.}\}$$

describe collective processes with account taken of spatial correlations.

$$Q(x,x')=\frac{\omega_{s(a)}}{2\pi\hbar c^2N_1^2}\sum_{j\in V_1(x)}\sum_{k\in V_1(x')}\{A_{s0}{}^-(x_j)A_{a0}{}^-(x_k)\times\exp[i(2\omega_p-\omega_s-\omega_a)t]+\text{h.c.}\}$$

describe four-wave parametric-interaction processes,

$$R_3(x)=\frac{1}{2N_1}\sum_{j\in V_1(x)}\sigma_3{}^j$$

is the density of half the difference of the populations, T_1 and T_2 are the longitudinal- and transverse-relaxation times,

$$\frac{1}{T_0{}^2}=\frac{4\pi|g|^2}{\hbar\omega_{s(a)}}N_V$$

gives the characteristic time of interaction between the atom and the field,

$$S_0(x)=(^1/_2-R_3(x))/N_1,\; A_0(x)=(^1/_2+R_3(x))/N_1,$$

describe spontaneous processes, $\zeta(x) = \cos qx$ is the parameter of the coupling of the Stokes and anti-Stokes fields ($q = (2\mathbf{k}_p - \mathbf{k}_s - \mathbf{k}_a)_x$ is the detuning along the x axis). For simplicity, Eq. (8) was derived assuming longitudinal pumping.

The system (8) must be supplemented with initial and boundary conditions. If the initial state of the system is assumed unexcited and uncorrelated, and only waves traveling from left to right are taken into account, these conditions take the form:

$$\begin{aligned} &n_{s(a)}(x, x', t=0)=F_{s(a)}(x, x', t=0)\\ &\quad=S(x, x', t=0)=A(x, x', t=0)=0, \\ &R_3(x, t=0)=-^1/_2,\; n_{s(a)}(x, 0, t)=n_{s(a)}(0, x, t)=0). \end{aligned} \tag{9}$$

The system (8) cannot be solved analytically. We present the results of a numerical solution, which we compare with results of the semiclassical[1] and mode[5,6] theories. Figure 1 shows plots of the number of scattered photons and of the density of the population half-difference versus the normalized time, at the end of a cylindrical sample $L = 1$ cm long. As shown in Refs. 1 and in Refs. 5 and 6, allowance for the propagation effects leads to the onset of amplitude–phase modulation of the scattering pulses. A modulation of just this kind takes place in our case.

The spatial distribution of the amplitudes of the scattered waves at a fixed instant of time is shown in Fig. 2. As seen from the figure, the distribution is inhomogeneous, but in contrast to the semiclassical and mode theories it is oscillatory. The spatial distribution of the population half-difference is likewise inhomogeneous and oscillatory. The reason for this difference from the aforementioned models is that here we take into account also the presence of spatial correlation between the emitters (the mathematical cause is the simultaneous dependence of $n_{s(a)}$ on x and x').

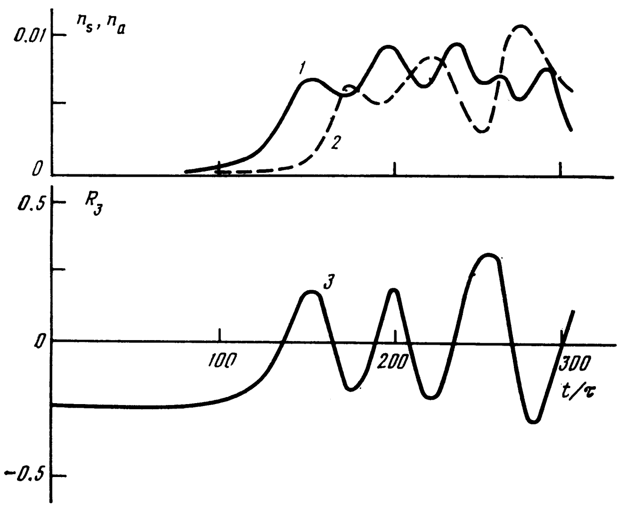

FIG. 1. Temporal evolution of cooperative Raman scattering at the end of a cylindrical sample: $\tau = 3\cdot10^{-11}$ s, $\tau_c = 4\cdot10^{-11}$ s, $T_2 = 10^{-9}$ s, $q = 8.7/L$, $R_3(x, t=0) = -0.5\exp[-(x-L/2)^2/0.32]$. 1—$n_s(L, L, t/\tau)$, 2—$n_a(L, L, t/\tau)$, 3—$R_3(L, t/\tau)$.

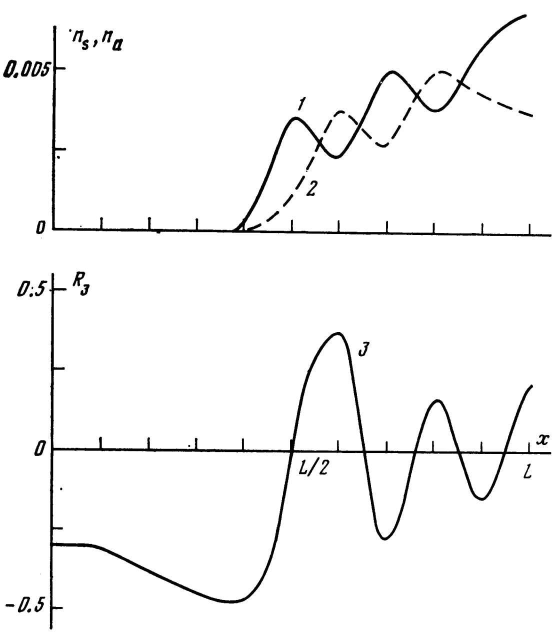

FIG. 2. Spatial distributions of the intensities of the scattered fields at a fixed instant of time $t_1 = t/\tau = 240$. 1—$n_s(x, x, t_1)$, 2—$n_a(x, x, t_1)$, 3—$R_3(x, t_1)$.

The degree of spatial correlation between the scattered fields, which are present in the small volume V_1 near the point $x' = L/2$ at a fixed instant of time, and fields having the same frequencies in neighboring sections, is shown by the functions $n_s(x, x' = L/2, t)$ and $n_a(x, x' = L/2, t)$. As seen from Fig. 3, a strong spatial correlation exists, is substantial in the vicinities of the point $x = L/2$, and decreases in oscillatory manner with increase of x. To the left of the point $x = L/2$ the correlation is insignificant, since only waves traveling from left to right are taken into account.

The theory developed permits investigation of the question of the influence of nonzero initial correlations on the dynamics of the evolution of the process in various regimes. This is a very important question, since the cooperative and stimulated Raman scattering differ in principle from each other, so that ways of identifying CRS in experiment can be indicated. This question was considered analytically for the linear stage of the evolution of the process, and by numerical methods in the general case.

Using the method of separating the spatial variables, we can represent the correlation function for the Stokes field component, during the linear stage when

$$|R_3(x, t)-R_3(x, 0)|\ll|R_3(x, 0)|,$$

in the form:

$$n_s(x, x', t)\propto\langle\mathbf{A}_s{}^+(x, t)\,\mathbf{A}_s{}^-(x', t)\rangle$$
$$=\left(\frac{2\pi c}{\omega_s}\frac{N}{V}\right)^2|g|^2\int_0^x dy\int_0^{x'} dz\, G_p{}^*(t, x-y)\,G_p(t, x'-z)\,S(x, x', 0). \tag{10}$$

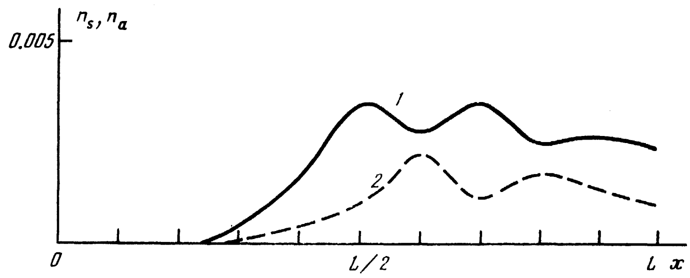

FIG. 3. Spatial distributions of correlation functions at a fixed instant of time $t_1 = t/\tau = 240$. 1—$n_s(L/2, x, t_1)$, 2—$n_a(L/2, x, t_1)$.

where $G_p(t, x-y)$ is the Green function of the linear problem (5) with (6) substituted. Putting $S(x,x',0) = |S(0)|^2\delta(x-x')$ it is easy to obtain from (10) the following expression for the correlation function:

$$n_s(x,x',t) \propto \int_0^x dy \exp\{-\alpha[(t-(x-y))+(t-(x'-y))]\}$$
$$\times\left[\frac{t-(x-y)}{2\beta\zeta(x-y)}\frac{t-(x'-y)}{2\beta\zeta(x'-y)}\right]^{1/2} I_1(2[2\beta\zeta(x-y)(t-(x-y))]^{1/2})$$
$$\times I_1(2[2\beta\zeta(x'-y)(t-(x'-y))]^{1/2})\theta(t-(x-y))\theta(t-(x'-y)). \tag{11}$$

Here $\alpha = \tau/2T_2$, t and x are the normalized time and coordinate ($t \to t/\tau$, $x \to x/L$), $\tau = L/c$, $\beta = 2\pi|g|^2/\hbar\omega$, and I_1 is a Bessel function of imaginary argument. Using the asymptotic representation of the functions I_1 for times on the order of the time of flight of the photon through the sample ($t \approx 1$), it is easy to obtain the following expression for the integral in (11):

$$n_s(x,x',t) \sim e^{-\alpha(x-x')}\left\{\frac{x-x'}{4\alpha^2}[1-e^{-2\alpha t}(2\alpha t+1)]+\frac{1}{4\alpha^3} - e^{-2\alpha t}\left(\frac{1}{4\alpha^3}+\frac{t}{2\alpha^2}+\frac{t^2}{2\alpha}\right)\right\}.$$

For cooperative Raman scattering, when $\alpha \ll 1$, we have consequently

$$n_s(x,x',t) \sim t^2[x-x'+t+1/2\alpha], \tag{12}$$

and in the case of stimulated Raman scattering, when $\alpha \gg 1$,

$$n_s(x,x',t) \sim e^{-\alpha(x-x')}(x-x'+1/\alpha)/4\alpha^2. \tag{13}$$

It follows thus from (12) and (13) that in the case of cooperative Raman scattering the correlation function at times $t \approx 1$ involves the entire volume of the sample, whereas in the case of stimulated Raman scattering the characteristic dimension of the correlation region for times $t \approx 1$ is $L_c \approx cT_2$.

The numerical-experiment results are shown in Figs. 4 and 5. Investigations have shown that in the case of stimulated Raman scattering the initial nonzero correlations increase rapidly with time, and the well-pronounced maxi-

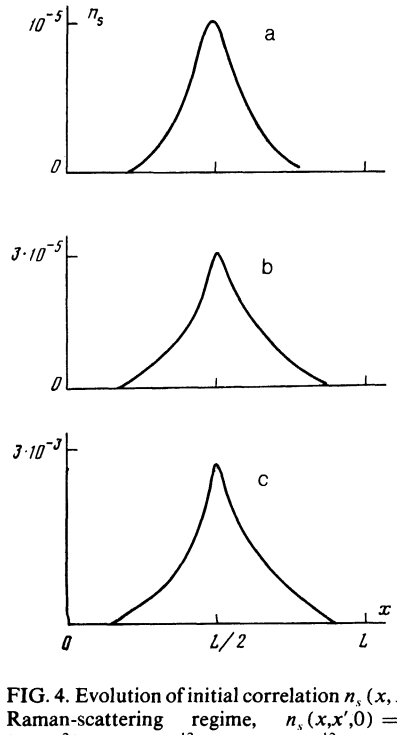

FIG. 4. Evolution of initial correlation $n_s(x, L/2, t/\tau)$ in the stimulated-Raman-scattering regime, $n_s(x,x',0) = 10^{-5} \exp[-(x-x')^2/2\cdot10^{-2}]$, $\tau = 3\cdot10^{-12}$ s, $\tau_c = 3\cdot10^{-12}$ s, $T_2 = 3\cdot10^{-13}$ s. a—$t=0$; b—$t/\tau = 1.2$; c—$t/\tau = 2$.

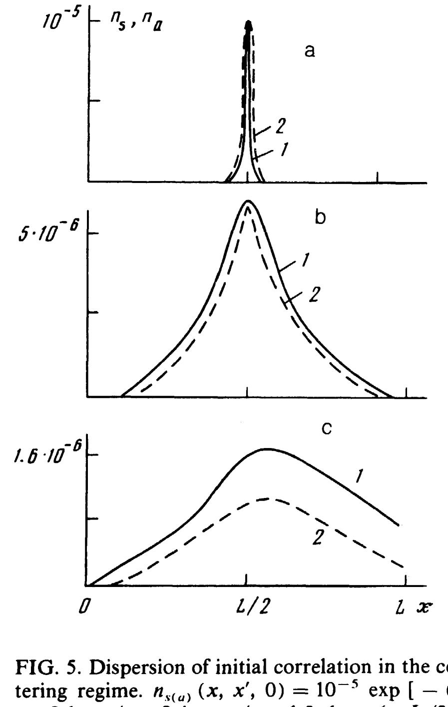

FIG. 5. Dispersion of initial correlation in the cooperative-Raman-scattering regime. $n_{s(a)}(x, x', 0) = 10^{-5} \exp[-(x-x')^2/2\cdot10^{-4}]$. a—$t=0$, b—$t/\tau = 0.4$; c—$t/\tau = 1.2$. 1—$n_s(x, L/2, t/\tau)$, 2—$n_a(x, L/2, t/\tau)$.

mum and the spatial centering of the plot are preserved. In the case of combined Raman scattering, the initial small correlation spreads out rapidly in space, disperses, and subsequently has no effect whatever on the evolution of the process.

The main advantage of the proposed theory is that it can yield, for the first time ever, a system of quantum equations describing the spatiotemporal development of cooperative scattering with an explicit allowance for propagation effects. It is possible to investigate in this theory the spatial distributions of the scattered-field intensities and the correlation functions. The analysis presented leads to the conclusion that the correlation properties of cooperative and stimulated Raman scattering are fundamentally different.

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