

Cooperative Raman scattering by a concentrated system of atoms

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A general solution is derived for the problem of the cooperative spontaneous Raman scattering of light by a concentrated system of two-level atoms. The given-field approximation is valid only if the number of atoms in the system, M , is sufficiently small. If M exceeds a certain critical M_{cr} , qualitative changes occur in the cooperative Raman scattering. The results derived here also apply to an extended system of atoms which can be described in the spatially homogeneous approximation. Possibilities for constructing a theory of cooperative Raman scattering incorporating effects of the field propagation in the medium are discussed.

Cooperative Raman scattering of light was predicted by Rautian and Chernobrod¹ and has been observed experimentally by Pivtsov *et al.*² The theory which has been constructed for this effect is based on the "given-field approximation"¹⁻⁶; i.e., it is assumed that the intensity of the Raman components of the scattered light is significantly lower than that of the incident laser light.

In the present paper we show that in the case of a concentrated system of atoms and also in the case of an extended system which can be described in the spatially homogeneous approximation¹ the given-field approximation is valid only if the number of atoms in the atomic subsystem, M , is sufficiently small. If M exceeds a certain critical $M_{cr} \approx \pi/J$ (J is the constant of the two-photon coupling of the light with an atom), qualitative changes occur in the behavior of the field-plus-atoms system, and the cooperative Raman scattering can no longer be treated as superradiance of Raman components in a given laser field.¹⁻⁶

If the spatially homogeneous approximation is not valid, and effects of the field propagation in the medium can play an important role, analysis of the cooperative Raman scattering becomes considerably more complicated. In the final section of this paper we show that the problem of cooperative Raman scattering in an extended system of atoms can be formulated in a classical two-dimensional model of statistical physics: the so-called six-vertex model.^{7,8}

1. FORMULATION OF THE PROBLEM

A laser pulse with a carrier frequency ω_L is incident on a system containing M two-level atoms which have a dipole-forbidden transition $1 \rightarrow 2$ with a frequency ω_{12} . The scattered field contains an unshifted component at the frequency ω_L and two shifted (Raman) components: a Stokes component with a frequency $\omega_S = \omega_L - \omega_{12}$ and an anti-Stokes component with a frequency $\omega_{AS} = \omega_L + \omega_{12}$. Furthermore, some of the incident pulse is not scattered and forms a transmitted laser beam.

We assume that all the atoms are in a sphere of radius r_0 which is much smaller than the typical wavelength of the light, λ . The existence of the small parameter $r_0/\lambda \ll 1$ makes it possible to expand the field operators in spherical harmonics⁹ and to retain only the electric-dipole harmonic with an angular momentum $j = 1$ when the field interacts with the ensemble of atoms. If levels 1 and 2 are nondegenerate, a specification of the polarization of the incident light fixes the second quantum number of the dipole photons, the angular-momentum projection m . We assume that the incident light

is linearly polarized. Choosing a quantization axis for the atoms along the field polarization direction, we then have $m = 0$.

We distinguish three intervals along the frequency axis: a laser interval, a Stokes interval, and an anti-Stokes interval, which are centered on the points ω_L , ω_S , and ω_{AS} , respectively, and which have width $\Delta \sim \tau_{in}^{-1}$, where τ_{in} is the length of the incident of the pulse. We use the indices L , S , and AS to specify that the dipole-photon operators $b(\omega) \equiv b(\omega, j = 1, m = 0)$ belong to one of these intervals. In the limit $\Delta \ll \omega_{12}$, all three harmonics are independent; i.e., the field operators with different indices commute with each other. We combine the field operators into an isotopic triplet

$$b_\sigma = \begin{pmatrix} b_{AS} \\ b_L \\ b_S \end{pmatrix} \quad (1)$$

with the commutation relations

$$[b_\sigma(k), b_{\mu^+}(q)] = 2\pi\delta_{\sigma\mu}\delta(k-q), \quad (2)$$

where the isospin variable σ (below we will say simply "spin variable") takes on three possible values: $\sigma = AS, L, S$.

The system of two-level atoms is described by the spin operator

$$R = \sum_{a=1}^M r_a = (R^x, R^y, R^z), \quad (3)$$

$$R^\pm = R^x \pm iR^y, \quad [R^i, R^j] = \epsilon^{ijk}R^k,$$

where the spin operators r_a ($r = \frac{1}{2}$) correspond to the a th atom.

In terms of the operators $b_\sigma(k)$ the Hamiltonian of the system consisting of the dipole photons and the atoms is

$$H = \int \frac{dk}{2\pi} \left\{ kb_{\sigma^+}(k) b_\sigma(k) - \int \frac{dk'}{2\pi} b_{\sigma^+}(k) [\sigma_{\sigma\sigma^+}^+ R^- + \sigma_{\sigma\sigma^+}^- R^+] b_{\sigma'}(k') \right\}, \quad (4)$$

where a repeated spin index implies a summation, and the 3×3 matrices

$$\sigma^+ = \begin{bmatrix} 0 & J_{AS} & 0 \\ 0 & 0 & J_S \\ 0 & 0 & 0 \end{bmatrix}, \quad \sigma^- = \begin{bmatrix} 0 & 0 & 0 \\ J_{AS} & 0 & 0 \\ 0 & J_S & 0 \end{bmatrix} \quad (5)$$

act in the spin space of the field. The operators representing the total number of particles,

$$N = \int \frac{dk}{2\pi} b_{\sigma^+}(k) b_{\sigma}(k), \quad (6)$$

and the z component of the spin of the field-plus-atoms system,

$$S^z = R^z + \int \frac{dk}{2\pi} b_{\sigma^+}(k) \sigma_{\sigma\sigma'}^z b_{\sigma'}(k), \quad (7)$$

where

$$\sigma^z = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{bmatrix},$$

commute with the Hamiltonian. We have accordingly omitted from expression (4) the terms $\omega_L N$ and $\omega_{12} S^z$, and we are placing the origin of the particle energy scale at the center of the corresponding frequency interval. Since the length of the incident pulse satisfies the inequality

$$\omega_L \tau_{in} \gg 1, \quad (8)$$

by assuming that again for the components of the scattered light this relation is not seriously violated we can extend the integration in (4), (6), (7) to infinite limits and ignore the frequency dependence of the coupling constants J_S and J_{AS} . Introducing

$$\varepsilon_{\sigma}(x) = \int_{-\infty}^{\infty} \frac{dk}{2\pi} b_{\sigma}(k) e^{ikx}.$$

we can rewrite the Hamiltonian of the problem as

$$H = - \int_{-\infty}^{\infty} dx \left[i \varepsilon_{\sigma^+}(x) \frac{\partial}{\partial x} \varepsilon_{\sigma}(x) + \delta(x) \varepsilon_{\sigma^+}(x) (\sigma_{\sigma\sigma'}^+ R^- + \sigma_{\sigma\sigma'}^- R^+) \varepsilon_{\sigma'}(x) \right]. \quad (9)$$

The problem of the Raman scattering of light by a concentrated system of atoms in the dipole ($r_0/\lambda \ll 1$) resonant [inequality (8)] approximation is thus described by an effectively one-dimensional quantum field theory. This type of one-dimensionalization is characteristic of several problems (e.g., the theory of magnetic alloys^{10,11} and the theory of Dicke superradiance¹²) in which a field interacts with a point impurity. In optical problems the role of an impurity is played by an atom or an ensemble of atoms concentrated in a small volume. In contrast with the theory of magnetic alloys, where the entities of interest are the equilibrium (thermodynamic) properties of a particle-plus-impurity system, the physical formulation of the problem of cooperative spontaneous Raman scattering corresponds to a many-body scattering problem, in whose initial state there are only laser photons. Consequently, the problem can be solved exactly even if model (9) is not completely integrable.

In the case of so-called resonant Raman scattering, with $J_{AS} \ll J_S$, the anti-Stokes component can be eliminated from consideration. It is sufficient for this purpose to replace the σ matrices (5), (7) by the corresponding Pauli matrices and to treat the field operators as constituting an isotopic doublet with a spin variable $\sigma = L, S$.

Model (9) is also used to describe cooperative Raman scattering in an extended system of atoms if the variations in the "slow" field amplitudes over the length of the sample can be ignored and if field-propagation effects in the medium are also ignored.¹ In such a case the atomic subsystem is again described by a total-spin operator R , while the variable x takes on the meaning of the coordinate along the axis of the sample. Consequently, all the results derived in the theory of cooperative Raman scattering for an extended system of atoms under conditions conforming to the spatially homogeneous approximation can thus be extracted from the results derived below for a concentrated system of atoms.

2. ONE-PARTICLE SCATTERING PROBLEM

Before we take up the solution of the general many-body scattering problem, we will examine the one-particle problem, in which a photon with a wave vector k ($|k| \equiv k = \omega - \omega_L$) is propagating along the z axis and is scattered by an m -fold-degenerate system of atoms at the point $z = 0$. The initial state of the field-plus-atoms system is

$$|In\rangle = \left[\frac{(M-m)!}{m! M!} \right]^{1/2} a_{k^+} (R^+)^m |0\rangle, \quad (10)$$

$$\langle In|In\rangle = 1,$$

where

$$a_{k^+} = \int dz \varphi(z) e^{ikz} a^+(z),$$

and $\varphi(z)$ is some arbitrary (normalized) wave function. The vacuum state is defined as the state in which there are no particles, and all of the atoms are in the ground state:

$$R^- |0\rangle = 0, \quad R^z |0\rangle = -\frac{M}{2} |0\rangle.$$

In other words, we are adopting a vector corresponding to the smallest eigenvalue of a completely symmetric (with a spin $R = M/2$) irreducible representation of the $SU(2)$ group, which acts in the state space of the atomic subsystem, as the vacuum state of the ensemble of atoms. If the vacuum state is the vector corresponding to the smallest eigenvalue in some arbitrary irreducible representation with a spin R , it is sufficient to replace M , the number of atoms, by $M_{\text{eff}} = 2R$ in all of the formulas.

Using an expansion of the operator a_k in spherical harmonics with a center at the point $z = 0$, we write it as the sum of two terms:

$$a_{k^+} = \tilde{a}_{k^+} + \left(\frac{\sigma_1}{S_0} \right)^{1/2} b_{L^+}(k), \quad (11)$$

where the operator \tilde{a}_k incorporates all the harmonics with angular momenta $j \geq 2$ which do not interact with the impurity (with the ensemble of atoms), $\sigma_1 = 3\pi/\omega_L^2$ is the impact area of the dipole particles,¹³ and S_0 is the cross-sectional area of the incident light beam. Expression (11) converts our original problem, (10), to the task of solving an auxiliary problem of the scattering of a dipole particle:

$$|in\rangle = b_{L^+}(k) (R^+)^m |0\rangle. \quad (12)$$

Its final state is evidently given by

$$|out\rangle = S |in\rangle, \quad (13)$$

where the S matrix of the scattering of the particle by the

impurity is found by solving a one-particle Schrödinger equation with Hamiltonian (9). The result is¹⁰

$$S = \exp [i(\sigma^+ R^- + \sigma^- R^+)]. \quad (14)$$

In expression (14) and below it is convenient to treat the σ^\pm as operators which act on the spin index of the field operators $b_\sigma(k)$. Here we will write out only the relations which we will need in our own problem:

$$(\sigma^+)^2 b_{L^+} = (\sigma^-)^2 b_{L^+} = 0, \quad (15a)$$

$$\sigma^+ b_{L^+} = J_{AS} b_{AS^+}, \quad \sigma^- b_{L^+} = J_S b_{S^+}, \quad (15b)$$

$$\sigma^- b_{AS^+} = J_{AS} b_{L^+}, \quad \sigma^+ b_{S^+} = J_S b_{L^+}.$$

These relations follow from the matrix representation of the σ operators, (5).

We rewrite expression (14) as the series

$$S = \sum_{n=0}^{\infty} \frac{(i)^n}{n!} (\sigma^+ R^- + \sigma^- R^+)^n, \quad (16)$$

and we consider the square of the operator:

$$(\sigma^+ R^- + \sigma^- R^+)^2 = (\sigma^+)^2 (R^-)^2 + (\sigma^-)^2 (R^+)^2 + \sigma^+ \sigma^- R^- R^+ + \sigma^- \sigma^+ R^+ R^-. \quad (17)$$

State (12) is an eigenstate for operator (17). According to (15a), the first two terms vanish, while the third and fourth [see (15b)] are equivalent to the operator $(J_S^2 R^- R^+ + J_{AS}^2 R^+ R^-)$. We can thus write

$$(\sigma^+ R^- + \sigma^- R^+)^2 |in\rangle = (J_S^2 R^- R^+ + J_{AS}^2 R^+ R^-)^l |in\rangle, \quad (18)$$

where l is a natural number.

When a state containing only a laser photon is acted upon, the expression for the S matrix thus takes the simple form

$$S = A + (\sigma^+ R^- + \sigma^- R^+) C, \quad (19a)$$

$$A = \cos(J_S^2 R^- R^+ + J_{AS}^2 R^+ R^-)^{1/2}, \quad (19b)$$

$$C = i \frac{\sin(J_S^2 R^- R^+ + J_{AS}^2 R^+ R^-)^{1/2}}{(J_S^2 R^- R^+ + J_{AS}^2 R^+ R^-)^{1/2}}. \quad (19c)$$

Taking account of the harmonics which do not interact with the impurity, we find the following solution of the one-particle scattering problem:

$$|Out\rangle = \left[\frac{(M-m)!}{m! M!} \right]^{1/2} \left\{ a_{k^+} + \left(\frac{\sigma_1}{S_0} \right)^{1/2} [(A-1) b_{L^+}(k) + (J_S R^+ b_{S^+}(k) + J_{AS} R^- b_{AS^+}(k)) C] \right\} (R^+)^m |0\rangle, \quad (20)$$

where the terms of the sum correspond to the transmitted laser light and the three components of the scattered light, respectively.

The intensities of the components of the scattered light are found by averaging the corresponding density operator $\hat{I}_\sigma(x) = \varepsilon_\sigma^+(x) \varepsilon_\sigma^-(x)$ (there is no summation over σ) over the Out state:

$$I_L(x) = 4I_D(x) \sin^4 [g(m)/2], \quad (21a)$$

$$I_S(x) = I_D(x) J_S^2 (m+1) (M-m) \frac{\sin^2 [g(m)]}{g^2(m)}, \quad (21b)$$

$$I_{AS}(x) = I_D(x) J_{AS}^2 m (M-m+1) \frac{\sin^2 [g(m)]}{g^2(m)}, \quad (21c)$$

where

$$g(m) = [J_S^2 (m-1) (M-m) + J_{AS}^2 m (M-m+1)]^{1/2}. \quad (21d)$$

In expressions (21), $I_D(x) = (\sigma_1/S_0) I_0(x)$ is the intensity of the dipole component of the incident laser light, whose intensity is $I_0(x) = |\varphi(x)|^2$. The derivation of these expressions is based on the relations

$$A (R^+)^m |0\rangle = \cos [g(m)] (R^+)^m |0\rangle,$$

$$C (R^+)^m |0\rangle = i \frac{\sin [g(m)]}{g(m)} (R^+)^m |0\rangle, \quad (22)$$

$$R^- (R^+)^m |0\rangle = -m (M-m+1) (R^+)^{m-1} |0\rangle,$$

which are simple consequences of the algebra of spin operators \mathbf{R} .

An important point here is that we need to allow for the interference of the states of the transmitted and scattered laser light, as we can see from the fact that the following commutator is nonzero:

$$[a_k, b_{L^+}(k)] = \left(\frac{\sigma}{S_0} \right)^{1/2}. \quad (23)$$

Only when we incorporate (23) will the Out state be correctly normalized ($\langle Out|Out\rangle = 1$), so the scattering will be unitary. As usual in scattering theory,¹³ we are assuming here that the incident beam has a finite cross section and that the measurements of the scattered field are carried outside this cross section. For this reason, the nonvanishing commutator (23) need be taken into account only for the operators b_L which appear in the definition of the final state of the scattering problem, while the operators b_L which figure in the operator of a measurable physical quantity must be assumed to commute with the operators of the transmitted wave, a_k .

As we will see below, the region of parameter values of the problem which is of physical interest is

$$J_S M, \quad J_{AS} M \sim 1. \quad (24a)$$

The intensity of the scattered light is thus substantially different from zero only if the system of atoms is fairly highly excited ($m \sim M/2$). For macroscopically large values of M the terms

$$J_S^2 M, \quad J_{AS}^2 M \sim M^{-1} \ll 1 \quad (24b)$$

are negligibly small and can thus be omitted from expressions (21). As a result we find

$$I_L(x) = 4I_D(x) \sin^4 [u(m)/2], \quad (25a)$$

$$I_S(x) = \frac{J_S^2}{J_{AS}^2} I_{AS}(x) = I_D(x) \frac{J_S^2}{J_S^2 + J_{AS}^2} \sin^2 [u(m)], \quad (25b)$$

$$u(m) = [(J_S^2 + J_{AS}^2) m (M-m)]^{1/2}. \quad (25c)$$

If the scattering is by an unexcited atomic subsystem ($m = 0$), we find from (21)

$$I_L(x) = \frac{1}{4} I_D(x) J_S^4 M^2, \quad (26a)$$

$$I_S(x) = I_D(x) J_S^2 M. \quad (26b)$$

As expected, relations (26) reproduce the known result that the scattering for the unshifted and shifted components is respectively coherent ($\propto M^2$) and incoherent ($\propto M$). This

conclusion was reached in Ref. 9 by taking an average of the intensity of the scattered light over the random (for each atom) phases of the coupling constant J_S . In the present paper we are assuming that the phase factors are incorporated in the definition of the atomic operators r_a^\pm ; this assumption does not alter the commutation relations for either these operators or the total-spin operators R^\pm . In our approach, relations (26) are thus a natural consequence of the algebra of spin operators.

3. MANY-BODY SCATTERING PROBLEM

The shape of the incident laser pulse plays a completely insignificant role. This property follows from the fact that the S matrix is independent of the momentum of the particle,¹⁰ and it evidently persists in the many-body case. The theory of cooperative Raman scattering can thus be constructed for an incident pulse of arbitrary shape.

In order to avoid extraneous and tedious intermediate calculations in the derivation of the physical observables of the scattered field below, we use the fact that the S matrix does not depend on the momentum of the particle in order to transform from a continuous description of the field to a discrete model. The initial state of the many-body scattering problem in terms of the node operators a_j, a_j^+ , with the commutation relation:

$$[a_j, a_i^+] = \delta_{ji},$$

is then written in the form

$$|In\rangle = \left[\frac{(M-m)!}{m! M!} \right]^{1/2} \left(\prod_{j=1}^N a_j^+ \right) (R^+)^m |0\rangle, \quad (27)$$

where the number of nodes, N , is equal to the average number of photons in the incident light:

$$N = \int_{-\infty}^{\infty} dt I_0(t).$$

Singling out the dipole harmonic in the node creation operator,

$$a_j^+ = \tilde{a}_j^+ + \left(\frac{\sigma_1}{S_0} \right)^{1/2} b_{L,j}^+,$$

we can again reduce our original problem, (27), to that of solving a set of auxiliary problems with an in-state:

$$|in\rangle = \left(\prod_j b_{L,j}^+ \right) (R^+)^m |0\rangle.$$

This state contains a certain number of dipole particles, between 0 and N . The dipole harmonic of the light is described by a triplet of fields with the commutation relations

$$[b_{\sigma,j}, b_{\mu,l}^+] = \delta_{\sigma\mu} \delta_{jl},$$

and the scattering of particle j by the impurity corresponds to the S matrix

$$S_{j0} = A + (\sigma_j^+ R^- + \sigma_j^- R^+) C,$$

where the operators σ_j act in the spin space of node j .

The final state of the auxiliary problem is thus given by the expression

$$|out\rangle = \left(\prod_j S_{j0} \right) |in\rangle.$$

Taking account of the field harmonics which do not interact with the impurity, we find

$$|out\rangle = \left[\frac{(M-m)!}{m! M!} \right]^{1/2} \left(\prod_{j=1}^N d_j^+ \right) (R^+)^m |0\rangle, \quad (28a)$$

where

$$d_j^+ = a_j^+ + \left(\frac{\sigma_1}{S_0} \right)^{1/2} [(A-1)b_{L,j}^+ + (J_S b_{S,j}^+ R^+ + J_{AS} b_{AS,j}^+ R^-) C]. \quad (28b)$$

The unitarity of the scattering of the dipole particles leads to a commutation relation for the d operators:

$$[d_j, d_i^+] = \delta_{ji}.$$

in the discussion below, the vanishing of the vacuum expectation value $\langle d_i^+ d_j \rangle$ makes it convenient to use

$$d_j d_i^+ = \delta_{ji} \quad (29)$$

as the unitarity condition.

In our problem of cooperative spontaneous Raman scattering, all of the incident particles are in the same spin state. This circumstance not only simplifies the expression for the S matrix but also eliminates many-body effects in the scattering which determine the thermodynamic properties of exchange models.^{10,11} Expression (28) corresponds to a sequential and independent scattering of particles by the impurity. As a result of the scattering, the state of the atomic subsystem changes; i.e., the particle of index $n+1$ is scattered by the impurity whose state arose as a result of the scattering of the preceding n particles. If relaxation processes in the system of atoms do not have time to destroy the "memory" of the scattered particles during the incident pulse, of length τ_{in} , the amplitude for the scattering of a particle will depend on its index. It is this circumstance which makes the scattering process "cooperative."

4. CALCULATION OF PHYSICAL OBSERVABLES; RECURRENCE RELATIONS

The physical characteristics of the scattered light are determined by averaging the corresponding node operators over the Out state (28). At this point we will simply evaluate the one-node correlation functions, which determine the intensity of the components of the scattered light:

$$\begin{aligned} I_\sigma(n+1) &= I_0(n+1) \langle Out | b_{\sigma,n+1}^+ b_{\sigma,n+1} | Out \rangle \\ &= I_0(n+1) \frac{(M-m)!}{m! M!} \\ &\quad \times \left\langle (R^-)^m \left(\prod_{j=1}^{n+1} d_j \right) b_{\sigma,n+1}^+ b_{\sigma,n+1} \left(\prod_{j=1}^{n+1} d_j^+ \right) (R^+)^m \right\rangle, \end{aligned} \quad (30a)$$

where n is the index of the node; $\sigma = L, S$, or AS ; and $I_0(n)$ is the intensity of the incident light. In the second of these equations we have made use of the unitarity of the d operators (29). Furthermore, we calculated the expectation value of the z component of the spin of the impurity (the population of the ensemble of atoms) which arises as a result of the scattering of n photons:

$$R_n = \frac{(M-m)!}{m!M!} \left\langle (R^-)^m \left(\prod_{j=1}^n d_j \right) R^+ \left(\prod_{j=1}^n d_j^+ \right) (R^+)^m \right\rangle. \quad (30b)$$

The angle brackets in (30) mean the expectation value over the vacuum state.

We switch to a continuous description by replacing the node index n by the function

$$n(t) = \int_{-\infty}^t dt' I_0(t'). \quad (30c)$$

Let us consider the expectation value

$$Q_{n+1}^m = \frac{(M-m)!}{m!M!} \langle (R^-)^m \hat{Q}_{n+1} (R^+)^m \rangle \quad (31a)$$

of the "Heisenberg" operator

$$\hat{Q}_{n+1} = \left(\prod_{j=1}^{n+1} d_j \right) \hat{q}_{n+1} \left(\prod_{j=1}^{n+1} d_j^+ \right), \quad (31b)$$

where \hat{q}_{n+1} is some arbitrary node operator ("Schrödinger operator"). Expanding the d operators at the far left and at the far right in (31) in accordance with definition (28b), and carrying out the corresponding calculations, in which we make use of expressions (22) and (23), we find the following recurrence relations for the quantities Q_n^m :

$$Q_{n+1}^m = Q_n^m + \frac{\sigma_1}{S_0} \left\{ -\sin^2[g(m)] Q_n^m + J_B^2(m+1)(M-m) \frac{\sin^2[g(m)]}{g^2(m)} Q_n^{m+1} + J_{AB}^2 m(M-m+1) \frac{\sin^2[g(m)]}{g^2(m)} Q_n^{m-1} \right\}, \quad (32)$$

where the function $g(m)$ is defined in (21d). Relations (32) are determined exclusively by the structure of the d operator; they do not depend on the form of the node operator. The structure of a q operator determines only the initial condition

$$Q_0(m) = Q_{n+1}^m |_{n=0},$$

which is required for a solution of the recurrence relations (32). As $Q_0(m)$ we evidently need to use the results of the solution of the one-particle problem for the intensities of the components of the scattered light, (21), or the expression

$$Q_0(m) = -M/2 + m \quad (33)$$

in determining the population of the atomic subsystem, (30b).

We restrict the analysis here to only those solutions of recurrence relations (32) which are suitably smooth— which vary only slightly over one lattice step, $n \rightarrow n \pm 1$, $m \rightarrow m \pm 1$. The initial conditions are evidently smooth functions of m . Accordingly, treating $Q(n, m)$ as a function of the continuous variables n and m , we switch from the recurrence relations to a Cauchy problem for the equation

$$\frac{\partial Q}{\partial n} - c(m) \frac{\partial Q}{\partial m} = 0, \quad (34a)$$

$$c(m) = \frac{\sigma_1}{S_0} [J_B^2(m+1)(M-m) - J_{AB}^2 m(M-m+1)] \frac{\sin^2[g(m)]}{g^2(m)} \quad (34b)$$

with the initial conditions

$$Q(n, m) |_{n=0} = Q_0(m). \quad (34c)$$

In the region of physical interest, (24), we can discard the small terms ($\sim M^{-1}$) from (34b) and rewrite the function $c(m)$ as

$$c(m) = \frac{\sigma_1}{S_0} \frac{J_B^2 - J_{AB}^2}{J_B^2 + J_{AB}^2} \sin^2[u(m)], \quad (35)$$

where $u(m)$ is defined in (25c). Correspondingly, we can use expressions (25a) and (25b) (without the factor I_D) as an initial condition.

To evaluate one-node correlation functions (30) it is thus sufficient to solve the Cauchy problem (34a), (34c), (35); restricting the discussion to scattering by an initially unexcited atomic subsystem for the discussion below, we set $m = 0$ in the solution. As a result we find

$$I_L(t) = 4I_D(t) \sin^4[u(m(t))/2], \quad (36a)$$

$$I_B(t) = \frac{J_B^2}{J_{AB}^2} I_{AB}(t) = \frac{J_B^2}{J_B^2 + J_{AB}^2} I_D(t) \sin^2[u(m(t))], \quad (36b)$$

$$R(t) = -M/2 + m(t), \quad (36c)$$

where the function $m(t)$ is determined implicitly by the equation

$$\frac{J_B^2 - J_{AB}^2}{J_B^2 + J_{AB}^2} \int_{-\infty}^t dt' I_D(t') = \int_0^{m(t)} \frac{dm'}{\sin^2[u(m')]} \quad (36d)$$

The expressions for the correlation functions in the many-body problem are completely identical to the corresponding expressions for the one-particle problem of the scattering by an m -fold-degenerate impurity, (25). The only manifestation of the many-body nature of the problem is the time dependence of m or, more precisely, the dependence of m on the number of photons which have been scattered by the time t .

The absence of many-body effects can also be seen in the structure of the multipoint correlation functions. For example, let us evaluate the two-point correlation function

$$Q(n, n') = \langle \hat{Q}_n \hat{Q}_{n'} \rangle, \quad (37)$$

where the Heisenberg operator \hat{Q}_n is given by (31b). The recurrence-relation method described above for the quantities $Q(n, n', m)$ leads to an equation for the function $Q(n, n', m)$:

$$\frac{\partial Q}{\partial n'} + \frac{\partial Q}{\partial n} - c(m) \frac{\partial Q}{\partial m} = 0, \quad (38a)$$

with the initial condition

$$Q(n, n', m) |_{n'=0} = Q_0(m) Q(n, m). \quad (38b)$$

The function $Q(n, m)$ here is evidently a solution of the Cauchy problem (34); i.e., the function $Q(n, m)$, which determines a single-point correlation function, plays the role of an initial condition in Cauchy problem (38) for the two-

point correlation function. Consequently, after a switch to a continuous description in which we make use of (30c), we can write the correlation function (37) as the product

$$Q(t, t') = Q(t)Q(t')$$

of one-point correlation functions. It is easy to see that to the extent that it is legitimate to transform from recurrence relations to a differential equation any multipoint correlation function can be broken up into a product of one-point correlation functions.

Finally, we note that only in the case $J_S > J_{AS}$ is there an important change in the population of the atomic subsystem in the course of scattering by an initially unexcited system of atoms. Consequently, we will use solutions (36) below only in this—physically interesting—case.

5. CRITICAL PHENOMENA

All the physical characteristics of the scattered field and the population of the atomic subsystem are determined by the one function $m(t)$, the equation for which, (36d), can be rewritten as

$$\frac{dm}{dt} = \frac{J_S^2 - J_{AS}^2}{J_S^2 + J_{AS}^2} I_D(t) \sin^2[u(m)], \quad (39a)$$

with the asymptotic condition

$$m(t \rightarrow -\infty) \rightarrow 0, \quad (39b)$$

The right side of Eq. (39a) is positive definite, so $m(t)$ is a monotonically increasing, bounded ($0 < m < M$) function.

The integrals of the corresponding components of the intensity of the scattered light, which determine the total numbers of laser photons, Stokes photons, and anti-Stokes photons,

$$N_s = \int_{-\infty}^{\infty} dt I_s(t),$$

can be put in the following form when we use (39):

$$N_s = \frac{J_S^2}{J_{AS}^2} N_{AS} = \frac{J_S^2}{J_S^2 - J_{AS}^2} \mu, \quad (40a)$$

$$N_L = \frac{J_S^2 + J_{AS}^2}{J_S^2 - J_{AS}^2} \int_0^{\mu} dm \operatorname{tg}^2\left(\frac{u(m)}{2}\right), \quad (40b)$$

where $\mu = m(t \rightarrow +\infty)$ is the maximum value of the population of the system of atoms, which, as we will see, is not always the same as the number of atoms, M .

In order to determine the time scale of the changes in the correlation functions, we switch to the function $u(t) = [(J_S^2 + J_{AS}^2) \times m(t)(M - m(t))]^{1/2}$ [see (25c)] in Eq. (39):

$$\begin{aligned} \frac{du}{dt} &= \operatorname{sgn}\left(\frac{M}{2} - m\right) \frac{M}{2} (J_S^2 - J_{AS}^2) I_D(t) \\ &\times \frac{\sin^2 u}{u} \left[1 - \left(\frac{u}{u_{\max}}\right)^2\right]^{1/2}, \end{aligned} \quad (41)$$

where

$$u_{\max}(M) = \frac{M}{2} (J_S^2 + J_{AS}^2)^{1/2} \quad (42)$$

is the maximum value of the function $u(m)$, which is reached at $m = M/2$. Replacing the function $I_D(t)$ in (41)

$$I_D = \frac{1}{\tau_{in}} \int_{-\infty}^{\infty} dt I_D(t) = \frac{N_D}{\tau_{in}},$$

where N_D is the total number of dipole photons in the incident flux, we find an expression for the time scale in the problem:

$$\tau_e = \frac{2\tau_{in}}{(J_S^2 - J_{AS}^2) N_D M}. \quad (43)$$

The parameter range $(J_S^2 - J_{AS}^2) N_D M \ll 1$ is thus of no physical interest.

The nature of the scattering process and the time evolution of the population of the system of atoms are determined completely by the quantity $u_{\max}(M)$.

1. We first consider the case $u_{\max} \ll 1$. Over the entire range the function $u(t)$ is much less than 1 in this case, so we can replace the function by the sine of its argument everywhere. It is the satisfaction of the inequality $u_{\max} \ll 1$ which makes the scattering efficiency low ($I_s \ll I_D$) and thus makes the given-field approximation valid. The solution of Eq. (36d) in this case is trivial:

$$m(t) = \frac{M}{1 + \exp[-2(t-t_0)/\tau_e]}. \quad (44a)$$

The expressions for the intensities become

$$I_L(t) = \frac{I_D}{64} (J_S^2 + J_{AS}^2)^2 M^4 \operatorname{sech}^4[(t-t_0)/\tau_e], \quad (44b)$$

$$I_s(t) = \frac{J_S^2}{J_{AS}^2} I_{AS}(t) = \frac{I_D}{4} J_S^2 M^2 \operatorname{sech}^2[(t-t_0)/\tau_e], \quad (44c)$$

where the parameter $t_0 = (\tau_e/2) \ln M$ characterizes the shift of the maximum values of $I_s^{\max} = \max I_s(t)$ after the initial time $t = 0$. Expressions (44) have been written here for the case of a step-shaped incident pulse: $I_D(t) = I_D \theta(t)$. With $J_{AS} = 0$ the expression for the Stokes component of the radiation becomes the same as the known results of the theory of cooperative Raman scattering in the given-field approximation.¹ The solutions have no oscillatory structure, regardless of the parameter values in the problem.

The intensity of the unshifted component of the scattered light is proportional to the fourth power of the number of atoms. This result applies specifically to a concentrated system; it cannot also be applied to the case of an extended system, for which there is no need to be particularly concerned about the behavior $I_L(t)$, since conservation of the number of photons tells us that we have

$$I_L(t) = I_0(t) - I_s(t) - I_{AS}(t),$$

and this relation does not hold for a concentrated system. In the scattering process the system of atoms becomes completely inverted; i.e.,

$$\mu \equiv m(t \rightarrow +\infty) = M, \quad (45)$$

and the values I_s^{\max} are reached at $m = M/2$. The total numbers of Stokes and anti-Stokes photons are proportional to the number of atoms, M [see (40a)], while the number of

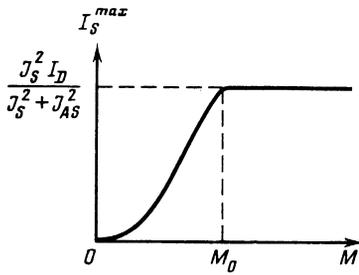


FIG. 1. Maximum intensity of the Stokes component of the scattered light, I_s^{\max} as a function of M .

laser photons, (40b), is proportional to M^3 :

$$N_L = \frac{(J_s^2 + J_{AS}^2)^2 M^3}{J_s^2 - J_{AS}^2 24}. \quad (46)$$

2. As the number of atoms in the system is increased, the growth of the maximum of the scattered-light intensities, $I_s^{\max}(M)$, deviates from a power law:

$$I_s^{\max} = \frac{J_s^2 I_{AS}^{\max}}{J_{AS}^2} = \frac{J_s^2}{J_s^2 + J_{AS}^2} I_D \sin^2 \left[\frac{M}{2} (J_s^2 + J_{AS}^2)^{1/2} \right], \quad (47a)$$

$$I_L^{\max} = 4I_D \sin^4 \left[\frac{M}{4} (J_s^2 + J_{AS}^2)^{1/2} \right]. \quad (47b)$$

Expression (47a) holds up to $M = M_0$:

$$u_{\max}(M_0) = \frac{M_0}{2} (J_s^2 + J_{AS}^2)^{1/2} = \frac{\pi}{2}.$$

For $M > M_0$, the increase in the maximum intensities of the shifted components comes to a halt (Fig. 1). These values are reached at the points $m = m_0^{(1,2)}$, where $m_0^{(1,2)}$ is found from the condition $u(M_0) = \pi/2$:

$$m_0^{(1,2)} = \frac{M}{2} \left\{ 1 \pm \left[1 - \left(\frac{M_0}{M} \right)^2 \right]^{1/2} \right\}. \quad (48)$$

The pulses thus become double-humped, and the distance between humps increases with increasing M . As before, the atomic subsystem becomes completely inverted in the course of the scattering ($\mu = M$), but the number of laser photons, N_L , is a complicated function of M [see (40b)].

3. A qualitative change in the scattering occurs at the value $M = M_{cr}$, where $u_{\max}(M_{cr}) = \pi$:

$$M_{cr} = \frac{2\pi}{(J_s^2 + J_{AS}^2)^{1/2}} = 2M_0. \quad (49)$$

For $M > M_{cr}$ the atomic system is not completely inverted, regardless of the intensities and regardless of the duration of

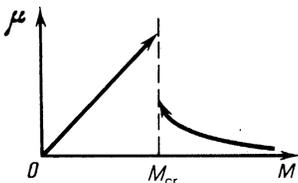


FIG. 2.

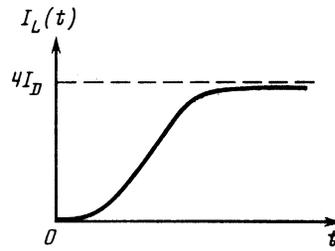


FIG. 3. Time evolution of the intensity of the unshifted component of the scattered light at $M > M_{cr}$.

the incident light. The maximum value of the function $m(t)$ is found from the condition $u(\mu) = \pi$:

$$\mu = \frac{M}{2} \left\{ 1 - \left[1 - \left(\frac{M_{cr}}{M} \right)^2 \right]^{1/2} \right\} < \frac{M}{2}. \quad (50)$$

For $M \gg M_{cr}$ the value of μ and, along with it, the values of N_S and N_{AS} [see (40a)] decrease in inverse proportion to the number of atoms, M : $\mu = M_{cr}^2/4M$. At the point $M = M_{cr}$, the function $\mu(M)$ is discontinuous (Fig. 2):

$$\mu(M_{cr}-0) - \mu(M_{cr}+0) = M/2. \quad (51)$$

The value $m = \mu$ is reached asymptotically in the limit $t \rightarrow +\infty$. In this case we have $u(t \rightarrow +\infty) \rightarrow \pi$, so the intensities of the shifted components of the scattered light have the shape of the pulse, and they vanish exponentially, while the intensity of the unshifted component tends toward the unitary limit¹³ (Fig. 3):

$$I_L(t \rightarrow +\infty) \rightarrow 4I_D = 4(\sigma_1/S_0)I_0. \quad (52)$$

Consequently, for $M > M_{cr}$ the intensity $I_L(t)$ increases from 0 at $t = 0$ to $4I_D$ in the limit $t \rightarrow +\infty$. The integral in (40b) diverges, and we find $N_L \rightarrow \infty$ as $M \rightarrow M_{cr} - 0$ (Fig. 4). Correspondingly, expression (47b) is valid only at $M < M_{cr}$.

6. EXTENDED SYSTEM OF ATOMS; SIX-VERTEX MODEL¹⁾

We turn now to an analysis of the problem of cooperative Raman scattering in an extended system of atoms, which has a length $l > \tau_c$, so the spatially-homogeneous approximation must be abandoned. We restrict the analysis to the case of resonant Raman scattering ($J_{AS} = 0, J_S \equiv J$) under conditions such that a one-dimensional model with unidirectional propagation of particles is valid.³⁻⁶ For this case

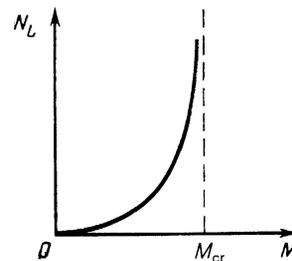


FIG. 4. Total number of unshifted scattered photons, N_L , versus M .

we rewrite the Hamiltonian of the problem in the form

$$H = - \int_{-\infty}^{\infty} dx \left\{ i e_{\tau}^{+}(x) \frac{\partial}{\partial x} e_{\tau}(x) + J \sum_{a=1}^M \delta(x-x_a) e_{\tau}^{+}(x) [\tau_{\tau}^{+} r_a^{-} + \tau_{\tau}^{-} r_a^{+}] e_{\tau}(x) \right\} \quad (53)$$

where $e_{\tau}^{+} = (E_L^{+}(x), E_S^{+}(x))$ is an isotopic spinor made up of the operators representing the "slow" amplitudes of the laser field and the Stokes field, τ^{\pm} are Pauli matrices, and $\{x_a\}$ are the coordinates of the atoms along the axis of the sample (the x axis; $0 < x_a < l$).

In the classical limit and in the limit of a continuous description of the resonant medium, the equations of motion for the dynamic variables of the field-plus-atoms system are the same as the equations which were studied in Refs. 3–5 in the given-field approximation and also in Refs. 14–17 by the method of the inverse scattering problem.¹⁸ In our approach, it is obvious that the model is integrable, since its Hamiltonian, (53), is the same (aside from some distinctions which are unimportant for the analysis below, namely, the Bose statistics of the particles and the multiple impurities) as the Hamiltonian of the anisotropic Kondo model, whose integrability was established by Wiegmann.¹⁹ We can thus apply to cooperative Raman scattering several elegant mathematical techniques which have been developed in the theory of integrable quantum-mechanical systems.^{7,8,10,11} The initial state of the scattering problem is of the form

$$|in\rangle = \left(\prod_{j=1}^N E_{L,j}^{+} \right) |0\rangle, \quad (54)$$

where again we are using the node description of the field, and we are assuming that there are no particles in the vacuum and that all the atoms are in the ground state. The scattering of the j th particle by the a th impurity is described by the S matrix^{19,10}

$$S_{ja} = \frac{1}{2} (1 \otimes 1 + \tau_j^z \otimes r_a^z) + \frac{b}{2} (1 \otimes 1 - \tau_j^z \otimes r_a^z) + c (\tau_j^+ \otimes r_a^- + \tau_j^- \otimes r_a^+), \quad (55)$$

where $b = \cos J$, $c = i \sin J$, and \otimes means the tensor product.

We consider a lattice consisting of N horizontal and M vertical lines, which correspond to photons and atoms, re-

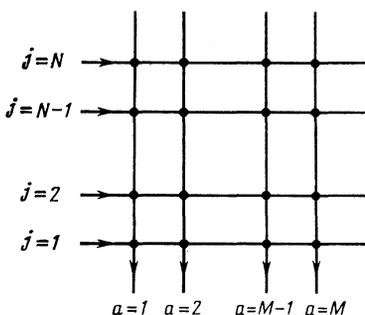


FIG. 5.

spectively (Fig. 5). We assume that the spin variables of the particles ($\tau_j = \pm 1/2$) and of the impurities ($r_a = \pm 1/2$) are given on the edges which are adjacent to the point of intersections of the lines, with which we associate S matrix (55). We represent the spin variables by arrows in the following way: An arrow directed to the right corresponds to the case in which the spin of the particle is up (a laser photon), while an arrow directed to the left corresponds to the case in which the spin of the particle is down (a Stokes photon). The direction of the arrows on the vertical lines is the same as the direction of the spin of the impurities. With the leftmost edges of the six-vertex model which has been described^{7,8} we associate the initial state of the system of particles (all the arrows point to the right), and with the bottom-most edges we associate the initial state of the atomic subsystem (all the arrows point down). The problem of the theory of spontaneous cooperative Raman scattering is to calculate either (a) the probabilities for the various directions of the arrows at the rightmost edges, in order to determine the state of the light which has passed through the medium, or (b) the probabilities for the various directions of the arrows at the uppermost edges, in order to determine the state of the atomic subsystem after the laser pulse has passed.

We treat the S matrix as a 2×2 matrix which acts in the spin space of the particles:

$$S_{ja} = \begin{bmatrix} \frac{1}{2} [(1+b) + (1-b)r_a^z] & cr_a^- \\ cr_a^+ & \frac{1}{2} [(1+b) - (1-b)r_a^z] \end{bmatrix}. \quad (56)$$

The scattering of the j th particle by the atomic subsystem is then described by a scalar product of matrices (56) (a monodromy matrix):

$$L_j = \begin{bmatrix} A & B \\ C & D \end{bmatrix} = \frac{1}{2} (1 + \tau_j^z) \otimes A + \tau_j^+ \otimes B + \tau_j^- \otimes C + \frac{1}{2} (1 - \tau_j^z) \otimes D. \quad (57)$$

When it acts on initial state (54), the monodromy matrix L_j reduces to the matrix

$$U_j = A + \tau_j^- \otimes C. \quad (58)$$

The final state of the scattering problem thus takes the form

$$|out\rangle = \prod_{j=1}^N [A E_{L,j}^{+} + C E_{S,j}^{+}] |0\rangle. \quad (59)$$

The structure of a U matrix is completely analogous to that of an S matrix in the case of a concentrated system, (19a), but now the operators A and C —in contrast with the corresponding operators in (19b) and (19c)—do not form a closed algebra. The effect is of course to substantially complicate the problem of calculating the correlation functions for the out state (59).

We introduce an S matrix which depends on some scalar parameter λ ,

$$S(\lambda) = \frac{1}{\text{ch } \eta} \begin{bmatrix} \sin(\lambda + i\eta r^z/2) & (i \text{sh } \eta) r^- \\ (i \text{sh } \eta) r^+ & \sin(\lambda - i\eta r^z/2) \end{bmatrix}, \quad (60)$$

and which is the same as (56) at the point $\lambda_0 = (\pi + i\eta)/2$, where $\cosh \eta = 1/\cos J$. For various values of λ , the ele-

ments of the monodromy matrix $L(\lambda)$, given in (57), form a closed algebra.^{7,8,10} The problem of cooperative Raman scattering thus reduces to one of calculating correlation functions for the state

$$|\text{out}, \{\lambda_j\}\rangle = \prod_{j=1}^N [A(\lambda_j)E_{L,j}^+ + C(\lambda_j)E_{S,j}^+] |0\rangle \quad (61)$$

and then taking the limit $\{\lambda_j\} \rightarrow (\pi + i\eta)/2$.

7. CONCLUSION

In this paper the ensemble of two-level atoms is described by the spin operator

$$\mathbf{R} = \sum_{\alpha=1}^M \mathbf{r}_{\alpha},$$

which is the sum of the spin operators of the individual atoms. This assertion is correct either for a concentrated system of atoms (Dicke geometry, $r_0/\lambda \ll 1$) or for an extended system of atoms, provided that the spatially-homogeneous approximation is valid, and provided that the length l of the system is much shorter than the length τ_c of the Stokes and anti-Stokes pulses. These two physically different situations are described by mathematically equivalent models, so all the results pertaining to the spatially homogeneous model²⁰ can be extracted from the results of the present paper.

A significant fraction of the experiments which have been carried out on cooperative Raman scattering can be described in the spatially homogeneous approximation. According to estimates by Zabolotskiĭ,⁵ for example, the effective length of the scattering volume in their experiments was $l \approx 10$ cm, while the length of the Stokes pulse was $\tau_c \approx (5-15) \cdot 10^{-9}$ s, so the parameter $l/c\tau_c$ ranged from 1/15 to 1/45.

In the same experiments, the maximum intensity of the Stokes component, I_S^{\max} , reached 70% of the intensity of the incident laser light, I_0 . From these figures we can estimate the value of the parameter $u_{\max}(M) = JM/2$ in the experiments of Ref. 5. According to (47a), in this case we have $\sin^2 u_{\max} = 0.7$, so u_{\max} was on the order of $\pi/3$. Consequently, in order to observe the critical phenomena in cooperative Raman scattering predicted in Ref. 20 and the present paper it is sufficient to increase the value of the parameter u_{\max} by a factor of only three from the value achieved in the experiments of Ref. 5.

In the case of a Dicke geometry ($r_0/\lambda \ll 1$), the cooperative Raman scattering acquires several interesting new features, which stem from interference between the transmitted laser light and the scattered unshifted component. The possibility of an experimental implementation of this phenomenon in the Dicke geometry requires a separate discussion, but a recent realization of the superradiance effect for a concentrated system of Rydberg atoms raises the hope that the same effect can be achieved in our case.

In the theory derived here for cooperative Raman scattering the physical quantities are determined through a solution of the recurrence relations (32). As we have shown here, a transformation from the recurrence relations to the differential equation (34a) results in the neglect of correlation effects. All of the multipoint correlation functions split up into a product of single-point correlation functions. It is

also obvious that Eq. (34a) is good for calculating only average statistical values of physical observables; information about their fluctuations is lost in the transformation from recurrence relations to a differential equation. In order to describe the correlation and statistical characteristics of the scattered light we thus need a more careful analysis of the recurrence relations (32) for single-point correlation functions and also of the recurrence relations for multipoint correlation functions, which can be derived easily by the method described here. One finds a hierarchy of recurrence relations, in which an n -point correlation function serves as an initial condition in the relation for an $(n+1)$ -point correlation function. We do not rule out the possibility that such an analysis will also make it possible to explain and describe the oscillatory structure which has been observed in the envelope of the Stokes pulses in several experiments.

If the length l of the system of atoms is greater than the length $c\tau_c$ ($l > c\tau_c$) scale of the problem, we must abandon the spatially homogeneous approximation and incorporate the effects of the field propagation in the medium. We have shown in this paper that the quantum-mechanical analog of the model which has conventionally been used in the semiclassical theory of cooperative Raman scattering^{3-6,14-17} is equivalent to an integrable Kondo model with an anisotropic exchange interaction.^{19,10} The corresponding multiparticle scattering problem can be formulated in the six-vertex model,^{7,8} so the final state of the scattered field, (61) can be determined; here the operators $A(\lambda)$ and $C(\lambda)$ form a closed algebra.^{7,8,10} Again in this case, the recurrence-relation method leads to closed expressions for the physical observables and to differential equation (34a), where the coefficient $c(m)$ is given by

$$c(m) = \frac{\langle (B)^{m+1} (C)^{m+1} \rangle}{\langle (B)^m (C)^m \rangle}. \quad (62)$$

Here the angle brackets mean the expectation value over the initial ("ferromagnetic") state of the atomic subsystem. Korepin²¹ has recently proposed a method for evaluating scalar products of the type

$$\left\langle \prod_{j=1}^m B(\lambda_j) \prod_{i=1}^m C(\mu_i) \right\rangle$$

with an arbitrary choice of rapidities $\{\lambda_j\}, \{\mu_i\}$. That development raises the hope that it will be possible to successfully complete the systematic derivation of an exact quantum-mechanical theory of the cooperative Raman scattering of an extended system of atoms even when the spatially homogeneous approximation is inapplicable.

¹⁾ The results of this section of the paper were derived in collaboration with A. I. Maĭmistov.

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