Stimulated Coherent Processes

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Scattering processes depend significantly on the prehistory of the scattering substance. In particular, coherent excitation of a scattering volume may considerably change the intensity and nature of the scattering. A quantum-mechanical theory is developed of light scattering by a system of impurity particles which has previously been excited to the collective state by coherent external sources. The energy spectrum of each particle contains two levels (R = 1/2). The case is considered when scattering in each elementary act involves at least two particles and the problem of determination of the scattering intensity is solved in the second order in perturbation theory. The possibility of phenomena of the stimulated Raman scattering type arising in scattering of coherent light is discussed. The scattering intensities of the first Stokes and anti-Stokes components are calculated ($\nu_1 \pm \nu_0$, where ν_1 is the scattered light intensity and ν_0 is the proper frequency of the impurity center).

OPTICAL induction and echo signals[1,2], and also stimulated optical induction signals[3] constitute coherent spontaneous responses of a system of weakly-interacting centers to the external action of laser sources. Since the intensity of the generated induction and echo signals is proportional to the square of the number of impurity centers (N^2), processes of this kind were named by Dicke[4] superradiance. Dicke reached the conclusion that superradiance has a purely spontaneous nature. On the other hand, stimulated processes, according to Dicke, have an intensity $\sim N$, as is indeed the case for superradiance processes with frequencies that are resonant with respect to the spectrum of the isolated center. The resonant superradiance process is the consequence of phased free precession of radiating multipoles, due to coherent and directional external laser excitation. On the other hand, if a system of particles, in a state in which it is capable of generating a superradiance signal in the regime of free precession with frequency ν_0, is subjected to an additional action of an external generator with frequency $\nu_1 \neq \nu_0$, then stimulated precession with frequency ν_1 is superimposed on the phased free precession of the radiating multipoles with frequency ν_0. As a result, the emission spectrum of such a system contains, in addition to the components of frequencies ν_1 and ν_0, also components with the combination frequencies $\nu_1 \pm \nu_0$. Since the free precession of the radiating multipoles occurs in phase in the coherent state, the intensity of the radiation at these frequencies, and particularly at the combination frequencies $\nu_1 \pm \nu_0$, is also proportional to N^2. In Dicke's terminology[4], this is the superradiance process (more accurately, superscattering), but in this case it has not a spontaneous but a stimulated character, since its intensity is also proportional to the square of the intensity of the scattered field.

The superscattering phenomenon is due to collective properties of quantum systems that are excited beforehand by coherent fields. In essence, a particle system excited to a collective state behaves with respect to the field that interacts with it like one tremendous "macromolecule," and the properties of this macromolecule can differ qualitatively from the properties of an individual particle.

Let us consider, for example, two particles with spin S = 1/2, with the interaction of each of them with the external fields described in the dipole approximation, which realizes transitions between the corresponding spin levels. In the collective state with respect to the interaction with the external fields, however, such a system behaves like one particle with spin 1 or 0, i.e., it already has a quadrupole moment. The emission (absorption) of a quantum of frequency $2\nu_0$ by such a system is the result of a simultaneous transition of both particles to the ground (excited) state. Consequently, when we deal subsequently with the scattering of light by a system of particles with spin S = 1/2, excited beforehand into a collective state, an important role will be played in the Hamiltonian of the interaction with the scattered fields not only by the dipole terms but also by the quadrupole terms.

We proceed to consider a system of N identical centers, each of which always has two energy levels, so that their interaction with the external field can be described in terms of an effective spin $R_j = {}^1/_2$ (j = 1, 2, ..., N)[4]. The collective state of such a system is described by the quantum numbers r and m, where r is the cooperation number and m is its projection on the quantization axis, which is the same for all particles ($r = {}^1/_2 N, {}^1/_2 N - 1, \ldots, |m| \leq r$). Raman scattering processes are considered within the framework of quantum electrodynamics as a result of an interaction of the scattered field of frequency ν_1 with the field of the zero-point oscillations[5]. Such processes in a system of N particles with effective spin R_j = 1/2 can proceed in each elementary scattering act with participation of at least two particles.

Assume that at the initial instant of time the system N of weakly-interacting centers is in a collective state with a state vector $\langle rms|$, where s is the aggregate of the quantum numbers describing the initial state of the field (scattered field plus the field of the zero-point oscillations). At succeeding instants of time, the system of particles will be in one of the collective states with wave function $\langle rm_k s_k|$. The Hamiltonian $\mathcal{H}_1$ of the interaction of the field with the particles contains both dipole terms $\mathcal{H}_1^D$, which are proportional to $R_\pm$, and quadrupole terms $\mathcal{H}_1^Q$, which are quadratic in the opera-

tors $R_\pm$. Here $R_\pm = R_1 \pm iR_2$ and R_1 and R_2 are the components of the effective spin of the entire system of particles

$$R_1 = \sum_{j=1}^{N} R_{1j}, \qquad R_2 = \sum_{j=1}^{N} R_{2j}. \tag{1}$$

The dipole part $\mathcal{H}_1^D$ of the interaction Hamiltonian describes single-particle transitions for the system, while the Hamiltonian $\mathcal{H}_1^Q$ describes two-particle transitions, wherein either one particle or two simultaneously take part, respectively, in each elementary emission (absorption) act.

The probability of transitions per unit time of the system of particles from the initial state $\langle rms|$ to the final state $\langle rm_k s_k|$ is obtained in second-order perturbation theory, and in the scattering process there are realized two types of intermediate states of the field[5]: 1) a quantum of frequency ν_1 (s = 1) is absorbed initially, there are no quanta in the intermediate state, and then a quantum ν_2 (s = 2) is emitted; 2) a quantum of frequency ν_2 is emitted initially, there are two quanta ν_1 and ν_2 in the intermediate state, and then on going to the final state there is absorbed a quantum ν_1. The quanta of frequency ν_2 pertain to the zero-point oscillations of the field with infinite frequency spectrum. In scattering processes, however, the frequencies ν_2 have certain definite values determined by the energy conservation law.

Let us consider, for example, all the processes that lead to a change of Δm by ± 1. The energy change corresponding to such transitions is equal to $h\nu_0$. Then the frequencies that take part in the scattering process satisfy the equation

$$\pm\nu_1 \mp \nu_2 = \nu_0. \tag{2}$$

All the processes with $\Delta m = \pm 1$ are described by crossing terms in the interaction Hamiltonian $\mathcal{H}_1$. Calculation in accordance with the same formulas of quantum-mechanical perturbation theory makes it possible to express the scattering intensity in a system of N particles in terms of the corresponding intensity for a system of two particles. In particular, for the scattering intensity at the combination frequency $\nu_1 + \nu_0$ we obtain the expression

$$I^{(\pm)} = \sum_{\sigma\xi\eta}{}' I^{(\pm)}_{\sigma\xi\eta} C^{rm}_{\sigma\xi\eta}. \tag{3}$$

At N = 2, the quantity $I^{(\pm)}$ is the intensity of scattering by a pair of isolated centers. The subscripts σ, ξ, η indicate that the scattering process corresponds to a transition described by the dipole part of the Hamiltonian $\mathcal{H}_1^D$ with R_σ ($\sigma = \pm$) and the quadrupole part with $R_\xi R_\eta$ ($\xi, \eta = \pm$), if R is taken to mean the effective spin of the two particles. The summation is over all the values of $\sigma, \xi, \eta = \pm$ corresponding to transitions with $\Delta m = \pm 1$. The coefficients $C^{rm}_{\sigma\xi\eta}$ are given by the formulas

$$\begin{aligned}
C^{rm}_{++-} &= (r-m+1)^2(r+m)^2(r-m)(r+m+1),\\
C^{rm}_{+-+} &= (r+m+1)^3(r-m)^3,\\
C^{rm}_{+--} &= (r-m+1)(r+m)(r-m+2)^2(r+m-1)^2,\\
C^{rm}_{-++} &= (r+m+1)^3(r+m)^3,\\
C^{rm}_{-+-} &= (r+m+1)(r-m)(r+m+2)^2(r-m-1)^2,\\
C^{rm}_{--+} &= (r+m+1)^2(r-m)^2(r-m+1)(r+m).
\end{aligned} \tag{4}$$

The quantities $I^{(\pm)}_{\sigma\xi\eta}$ are expressed in terms of the interaction constants $A^{\alpha}_{s\sigma}$ and $B^{\alpha}_{s\xi\eta}$ ($\alpha = \pm$), and also in terms of the occupation numbers n_s of the quanta of the field of the mode s. The general expression for $I^{(\pm)}_{\sigma\xi\eta}$ is

$$\begin{aligned}
I^{(\pm)}_{\sigma\xi\eta} = \frac{1}{8}\frac{\pi^2}{\hbar^4}\rho(\nu_k) n_1(n_2+1)\,|B^{+}_{2\xi\eta}A_{1\sigma} - [(\nu_{\sigma 0}-\nu_1)^{-1} + (\nu_{\xi\eta 0}-\nu_1)^{-1}]\\
+ B_{1\xi}A_{2\sigma}{}^{+}[(\nu_{\sigma 0}+\nu_2)^{-1} + (\nu_{\xi\eta 0}+\nu_2)^{-1}]\,|^2\delta(\pm\nu_0+\nu_2-\nu_1)h(\nu_1\pm\nu_0)
\end{aligned} \tag{5}$$

where $\rho(\nu_k)$ is the number of final states, and ν_{σ_0} and $\nu_{\xi\eta_0}$ are the frequencies of the transitions from the initial to the intermediate state, due respectively to the dipole part in $\mathcal{H}_1$ with R_σ or the quadrupole part with $R_\xi R_\eta$.

It is seen from (3) that at r = m = N/2 (all the particles are excited) the scattering intensity is given by

$$I^{\pm} = (I^{(\pm)}_{++-} + I^{(\pm)}_{+-+} + I^{(\pm)}_{+--} + I^{(\pm)}_{-++} + I^{(\pm)}_{-+-} + I^{(\pm)}_{--+})N^3. \tag{6}$$

On the other hand, if r = N/2 and m ≈ 0 (superscattering), then it is easy to verify that the scattering intensity becomes proportional to N^6.

The influence of the collective properties of a system of identical centers, caused by their interaction via the common radiation field, for the case when the elementary act of scattering with large intensity occurs on one isolated center (system of many-level particles) was investigated by a number of workers[6-8]. In[6,7] the processes of scattering and reflection of light were investigated on the basis of the balance equations. In[8], a classical theory of the influence of radiative interaction of atoms and molecules on these processes has been developed.

The role of the preliminary coherent excitation (PCE) in scattering processes was investigated by Kopvillem[9-11] who has shown, in particular, that PCE can be used to accelerate elementary particles. One of the main conclusions of these studies is that allowance for the radiative interaction or PCE leads to the appearance of a secondary emission component ~ N^2. Unlike the cases considered in[6-8], the appearance of a term ~ N^2 in the case of PCE does not impose additional limitations on the linear dimensions of the scattering and reflecting volumes.

We note in conclusion that the superscattering effect can be used to detect phenomena such as optical echo, and also stimulated optical induction[1-3]. Thus, if optical echo is excited in a system of multilevel particles (for example $Cr^{3+}:Al_2O_3$), then in the dipole approximation of single-particle scattering the intensity of the process at the frequency $\nu_1 \pm \nu_0$ (instants of time $t \geq \tau$, τ is the interval between the first and second exciting pulses) is given by

$$I_{\nu_1\pm\nu_0} = \frac{1}{16} I^{(0)}_{\nu_1\pm\nu_0} N^2 \frac{(\lambda_1 \pm \lambda_0)^2}{S}\sin^2(\theta_1 + \theta_{\text{eff}}), \tag{7}$$

where $I^{(0)}_{\nu_1\pm\nu_0}$ is the intensity of scattering by one isolated center, $\lambda_1 \pm \lambda_0$ is the wavelength corresponding to the frequency of the scattered light, S is the cross section area of the sample, and $\theta_1 + \theta_{\text{eff}} \approx \pi/2$. It is important to emphasize that this makes the frequencies of the detected and scattered signals different.

Let us estimate the ratio of the superscattering intensity (7) to the intensity of the corresponding scattering when no PCE is used. Let the optical echo be excited by two exciting pulses from a ruby laser ($\lambda_0 = 0.69\ \mu$), in analogy with the procedure used by Abella et al.[2]. The scattered field is light from a helium-neon laser with $\lambda_1 = 0.6328\ \mu$. If the number of scattering centers is $N = 10^{19}$, $S = 1\ cm^2$, and $\theta_1 + \theta_{eff} = \pi/2$, then the sought ratio is 4×10^{10}, which greatly increases the possibilities of experiments of this kind.

Let us call attention to the fact that the PCE can in principle be dispensed with. The coherence is produced in the system by the scattering of the coherent field ν_1 itself. Thus, if the scattered field is specified in the form of a pulse, then the frequency spectrum of the scattered field can contain the resonant frequency ν_0 and the field with this frequency brings the system of particles into a coherent state. This leads in turn to an increase of the intensity of scattering of all the nonresonant frequencies in the spectrum of the pulsed field. Such a process is indeed observed in stimulated Raman scattering (SRS).

We note finally that the second field combining with the incident field need not necessarily be the zero-point oscillations. This may be any periodic oscillation (acoustic waves, spin waves, etc.) In exactly the same way, the nature of the splittings of the impurity centers can also be arbitrary (orbital splitting, rotational and vibrational levels of the molecules, etc.).

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107