

monatomic spectroscopy of artificially obtained atoms of transuranium elements.

We note finally that the most suitable objects for the first investigations by this method are apparently even isotopes of alkaline-earth metals, in which one can use as resonant transitions the strong allowed transitions of the $n^1S_0 - n^1P_1$ type. The main characteristics of the cooling and capture process for three such isotopes are given in the table.

The power level required to saturate the resonant transitions of the isotopes in question is of the order of 100 mW at wavelengths that are quite attainable with tunable cw dye lasers. The laser-radiation line widths needed for the first experiment (much smaller than the natural width, for example on the order of 1 MHz) are likewise perfectly attainable in the cw regime. All this makes the new approach to laser spectroscopy, demonstrated in the present paper, fully realistic.

¹This force is called "gradient force" in^[6,7]. This designation should be regarded as less appropriate, since it indicates essentially only the formal method of obtaining the corresponding expression.

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Cooperative effect in Raman scattering of light

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A study is made of the influence of the interatomic correlation which arises in the process of emission of radiation on the spontaneous Raman scattering of light. It is shown that in the resonance scattering of a quasimonochromatic field, the shape of the Stokes component of the scattered pulse is identical with a superradiance pulse of the kind which appears in a medium of two-level atoms. If allowance is made for the anti-Stokes component, the scattering process becomes oscillatory.

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1. INTRODUCTION

Spontaneous emission from closely spaced atoms cannot be regarded as emission from isolated atoms because in the course of emission the atoms begin to interact with one another via the radiation field. The first quantum theory of the cooperative spontaneous emission from atoms was given by Dicke in^[1]. He showed that the rate of cooperative emission depends on the distribution of the phases of atomic oscillators and can vary

from γ_0 to $\gamma_0 N$, where γ_0 is the rate of the spontaneous decay of an excited state of an isolated atom and N is the number of atoms. Subsequently several authors^[2-4] studied the temporal and spatial characteristics of cooperative spontaneous emission of radiation from atoms in macroscopic regions. It was shown in particular^[2,3] that, in the quasiclassical approximation in the absence of an interatomic correlation at the initial moment, the radiation pulse has the shape of a hyperbolic secant on the time axis with a characteristic time $\tau_c \sim (\gamma_0 N/2)^{-1}$

and a delay relative to the initial moment amounting to $t_0 \sim \frac{1}{2}\tau_c \ln N$. Nagibarov and Kopvillem^[2] demonstrated that any process of boson emission is avalanche-like if the radiators interact via the field they create.

We shall consider the cooperative spontaneous Raman scattering of light which has the general properties of the cooperative spontaneous emission. For example, in the case of scattering of a quasimonochromatic pulse the process is described in the resonance approximation by the same time dependence as the collective spontaneous emission from two-level atoms and all the expressions can be obtained by replacing γ_0 with γ_R , where γ_R is the rate of transitions in isolated atoms under the Raman scattering conditions. In the nonresonance case, the cooperative emission is oscillatory (Sec. 4).

2. DERIVATION OF EQUATIONS

We shall consider the Raman scattering by atoms of a quasimonochromatic traveling wave of carrier frequency ω_{k_1} which is in the form of a step on the time axis, so that the rise time of the leading edge is shorter than all the other characteristic times of the problem. The scattering produces fields at the Raman Stokes and anti-Stokes frequencies. We shall adopt the following restrictions.

1) We shall use the quasiclassical approximation, i. e., we shall assume that the fields are classical and the behavior of atoms is described by the $\hat{\rho}$ matrix.

2) We shall confine our treatment to the one-dimensional approximation. We shall assume that the region occupied by atoms is in the form of a tube of length l and radius r such that $l \gg r$.

3) The delay time t_0 of a superradiance pulse [see Eq. (9)] is shorter than the atomic dephasing time T_2 . In this case the dephasing does not interfere with the appearance of an interatomic correlation.

4) The characteristic time of a superradiance pulse τ_c [see Eq. (9)] is greater than the transit time of a light signal through the tube $\tau_t = l/c$. This condition ensures that the interatomic correlation is established throughout the volume under consideration, and it allows us also to ignore the stimulated Raman scattering. In fact, the gain α of the Stokes component for gain times $t \ll T_2$ is related to τ_c as follows: $\alpha = l^{-1}(\tau_t/\tau_c)^{1/2}$. If $\tau_c \gg \tau_t$, we have $\alpha l \ll 1$, i. e., the gain in the length of the tube can be ignored.

5) The problem is assumed to be spatially homogeneous.

6) $|G_{1n}/\Omega_{1n}| \ll 1$, where $G_{1n} = E(k_1)d_{1n}/2\hbar$, $E(k_1)$ is the intensity of the scattered field, d_{1n} is the matrix element of the dipole moment, $\Omega_{1n} = \omega_{k_1} - \omega_{1n}$ is the detuning of the scattered-field frequency from resonance. This condition ensures the validity of the quasistationary approximation employed in Secs. 2 and 3.

7) $\tau_c^{-1} \ll |G_{1n}|^2/|\Omega_{1n}|$. This condition allows us to ignore the influence of time modulation of the refractive index.

The vector potential of the electromagnetic field will be assumed to be

$$A(z, t) = \sum_k \frac{1}{2} A(k, t) \exp(-i\omega_k t + ikz) + \text{c.c.},$$

where $k = k_1, \pm k_s$, and $\pm k_a$ are the wave factors of the strong field, Stokes component, and anti-Stokes component, respectively. We shall assume that $A(k_1, t) = \text{const}$, i. e., we shall ignore the change in the strong field. The scattered-light components will be described by a phenomenological equation for the field envelope inside the volume under consideration:

$$\frac{dA(k, t)}{dt} + \Gamma A(k, t) = -\frac{4\pi i c}{\omega_k} j(k, t), \quad (1)$$

where

$$j(k, t) = \frac{1}{V} \int dV j(r, t) e^{ikr}, \quad j(r, t) = \sum_{i,n} j_{im} P_{mi}(r, t),$$

$$P_{mi}(r, t) = \sum_i \rho_{mi}^i(r, t) \delta(r - r_i), \quad \Gamma_n = c/2l,$$

j_{im} is a matrix element of the current, ρ_{mi}^i is an element of the $\hat{\rho}$ matrix of an i -th atom. The function $j(k, t)$ is described by

$$j(k, t) = \frac{1}{V} \sum_{i,n} j_{im} P_{mi}(k, t), \quad P_{mi}(k, t) = \sum_i \rho_{mi}^i(t) \exp(ik \cdot r_i).$$

The interaction Hamiltonian will be taken in the form

$$H_{int} = -\frac{1}{c} \int dV A(r, t) \hat{j}(r, t). \quad (2)$$

We shall now give the system of equations for the elements of the collective $\hat{\rho}$ matrix of the spatially homogeneous problem in the interaction representation:

$$dN_i(t)/dt = -2 \operatorname{Re} \left[\sum_{k,n} i P_{1n}(k, t) g_{n1} A(-k, t) \right], \quad (3a)$$

$$dN_2/dt = 2 \operatorname{Re} \left[\sum_{k,n} i g_{2n} P_{n2}(k, t) A(-k, t) \right], \quad (3b)$$

$$\begin{aligned} & dP_{1n}(k, t)/dt + i(\omega_k - \omega_{n1}) P_{1n}(k, t) \\ &= -i g_{1n} N_{1n}(t) A(k, t) - i \sum_q P_{12}(-q, t) g_{2n} A(k+q, t), \end{aligned} \quad (3c)$$

$$\begin{aligned} & dP_{n2}(k, t)/dt - i(\omega_k - \omega_{n2}) P_{n2}(k, t) \\ &= i g_{n2} N_{2n}(t) A(k, t) + i \sum_q g_{n1} P_{12}(-q, t) A(k+q, t), \end{aligned} \quad (3d)$$

$$dP_{12}(-q, t)/dt = \sum_{k,n} [i g_{1n} P_{n2}(k, t) A(-k, t) - i P_{1n}(k, t) g_{n2} A(-k-q, t)], \quad (3e)$$

$$\sum_n N_n(t) = \text{const}. \quad (3f)$$

Here,

$$N_m = \sum_i \rho_{mm}^i, \quad g_{mn} = i\omega_{mn} d_{mn}/2\hbar c.$$

The summation in Eqs. (3a)–(3f) is carried out over all

the values of the wave vector \mathbf{k} and over all the intermediate energy levels of the atom.

Equations (1) and (3a)–(3f) form a closed system. We shall confine further discussion to the quasistationary approximation, i.e., we shall simplify Eqs. (1), (3c), and (3d) by dropping the derivatives with respect to time. This is permissible if the conditions 4), 6), and 7) above are satisfied. When this is done, Eqs. (1), (3c), and (3d) become algebraic and the quantities $A(k, t)$, $P_{1n}(k, t)$, and $P_{n2}(k, t)$ can be taken from these equations and substituted into Eqs. (3a), (3b), and (3e) so as to reduce the order of this system of equations. We shall omit simple but time-consuming steps and give the final equations:

$$dN/dt = -2g_s P_s + 2g_a P_a; \quad (4a)$$

$$dP_{s1}/dt = g_s N P_{s1}, \quad dP_{a1}/dt = -g_a N P_{a1}, \quad (4b)$$

where

$$N = N_1 - N_2, \quad P_{s,a} = |P_{12}(k_s, a - k_1)|^2 + |P_{12}(-k_s, -k_1)|^2,$$

$$g_{s,a} = \frac{8\pi c^2 \hbar}{V \Gamma_s \omega_{s,a}} |v_{s,a}|^2 |A(k_1)|^2,$$

$$v_s = \sum_n g_{2n} g_{n1} \left(\frac{1}{\omega_s + \omega_{n1}} + \frac{1}{-\omega_s + \omega_{n2}} \right),$$

$$v_a = \sum_n g_{2n} g_{n1} \left(\frac{1}{-\omega_a + \omega_{n1}} + \frac{1}{\omega_a + \omega_{n2}} \right).$$

The amplitudes of the Stokes and anti-Stokes components are now

$$A(k_s, t) = \frac{4\pi c^2 \hbar}{V \Gamma_s \omega_s} v_s A(k_1) P_{21}(k_s, -k_1), \quad (5a)$$

$$A(k_a, t) = \frac{4\pi c^2 \hbar}{V \Gamma_a \omega_a} v_a A(k_1) P_{12}(k_a, -k_1). \quad (5b)$$

3. RESONANCE APPROXIMATION

We shall assume that the frequency of the strong field ω_{k_1} is close to a transition between a level 1 and some intermediate level n , so that

$$|\omega_1 - \omega_{n1}| \ll |\omega_1 - \omega_{n2}|, \quad |\omega_1 + \omega_{n1}|.$$

Then, the solution of the system (4a)–(4b) can be obtained employing the resonance approximation, i.e., assuming that $g_s \gg g_a$ and ignoring the terms representing the anti-Stokes component in Eq. (4a); with the same accuracy we can assume that $(i\omega_{n2}/c)A(k_s, t) = E(k_s, t)$. Then, the system (4a)–(4b) becomes

$$dN/dt = -2g_s P_s, \quad (6a)$$

$$dP_{s1}/dt = g_s N P_{s1}, \quad (6b)$$

where

$$g_s = \frac{2\pi\omega_s |d_{2n}|^2}{\hbar V \Gamma_s} \left| \frac{G_{1n}}{\Omega_{1n}} \right|^2.$$

The system (6a)–(6b) contains an integral of motion

$$4P_{s1} + N^2 = R^2. \quad (7)$$

It follows from Eqs. (7) and (6a) that

$$dN/dt = -1/2 g_s (R^2 - N^2). \quad (8)$$

Equation (8) has the solution

$$N = -R \operatorname{th} \frac{t - t_0}{\tau_c}, \quad (9)$$

where

$$\tau_c = \frac{2}{g_s R}, \quad t_0 = \frac{\tau_c}{2} \ln \frac{R + N(0)}{R - N(0)}.$$

It follows from Eq. (4) that the Raman scattering intensity is $I_s = g_s \hbar \omega_s P_s / 2\sigma$, where σ is the cross section of the selected tubular region. Using Eqs. (7) and (9), we find that I_s is described by

$$I_s = \frac{g_s \hbar \omega_s}{2\sigma} R^2 \operatorname{sech}^2 \frac{t - t_0}{\tau_c}. \quad (10)$$

We shall consider the initial conditions corresponding to the absence of the interatomic correlation at the initial moment. Then, the initial intensity of the scattering into $k = \pm k_s$ modes is proportional to the number of particles at the level 1, so that the initial value of $P_s(t)$ will be taken to be $P_s(0) = 2N_1(0)$; we shall assume that $R = N_0$, where N_0 is the total number of atoms. If $N(0) \sim N_1(0) \sim N_0$, we find that $t_0 \sim \frac{1}{2} \tau_c \ln(N_0/2)$. In this case the solution (10) describes a pulse with a characteristic time $\tau_c \ll t_0$, which reaches its maximum at $t = t_0$.

The system (6a)–(6b) and its solution obtained on substitution $|G_{1n}/\Omega_{1n}| d_{2n} - d_{2n}$ are identical with the system of equations and its solution describing the cooperative spontaneous emission from two-level atoms.^[3] Therefore, the cooperative Raman scattering can be interpreted as the cooperative emission from effective dipoles and the magnitude of each dipole is $d = d_{2n} |G_{1n}/\Omega_{1n}|$.

4. NONRESONANCE CASE

We shall now consider the case of large detuning from resonance $|\omega_1 - \omega_{n1}| \sim |\omega_1 + \omega_{n1}|$. For the system (4a)–(4b) the integral of motion is

$$N^2 + 4(P_s + P_a) = R^2. \quad (11)$$

We shall now take P_a from Eq. (11) and substitute it into Eq. (4a):

$$dN/dt = 1/2 g_s (R^2 - N^2) - 2(g_s + g_a) P_s, \quad (12a)$$

$$dP_{s1}/dt = g_s N P_{s1}. \quad (12b)$$

Substituting in Eqs. (12a) and (12b) the value $g_a = g_s$ and going over to a second-order equation for N , we obtain

$$d^2 N/dt^2 = -1/2 g_s^2 (R^2 - N^2) N. \quad (13)$$

Equation (13) has the integral of motion

$$1/2 (dN/dt)^2 - 1/8 g_s^2 (R^2 - N^2)^2 = -1/8 g_s^2 a^2. \quad (14)$$

Using Eqs. (11) and (12a), we can express a^2 in terms of the initial values $P_a(0)$ and $P_s(0)$;

$$a^2 = 64P_s(0)P_a(0).$$

Taking dN/dt from Eq. (14), we obtain

$$dN/dt = \pm 1/2 g [(R^2 - N^2)^2 - a^2]^{1/2}. \quad (15)$$

Equation (15) is satisfied by the solution (see^[15])

$$N(t) = (R^2 - a)^{1/2} \operatorname{sn}(u, k), \quad (16)$$

where $\operatorname{sn}(u, k)$ is the elliptic Jacobi function,

$$u = (R^2 + a)^{1/2} \frac{gt}{2} + \int_0^{N(0)} \frac{dN}{[(R^2 - N^2)^2 - a^2]^{1/2}}, \quad k^2 = \frac{R^2 - a}{R^2 + a}$$

Equations (11), (12a), and (12b) give the following expressions for $P_s(t)$ and $P_a(t)$:

$$P_s(t) = 1/8 R^2 \operatorname{cn}^2(u, k) - 1/8 a \operatorname{sn}^2(u, k) + 1/8 (R^2 - a)^{1/2} \operatorname{cn}(u, k) \operatorname{dn}(u, k), \quad (17a)$$

$$P_a(t) = 1/8 R^2 \operatorname{cn}^2(u, k) - 1/8 a \operatorname{sn}^2(u, k) - 1/8 (R^2 - a)^{1/2} \operatorname{cn}(u, k) \operatorname{dn}(u, k). \quad (17b)$$

We can use Eqs. (5a) and (5b) to express the intensities of the Stokes and anti-Stokes components in terms of $P_{a,s}$:

$$I_{a,s} = (g\hbar\omega_{a,s}/2\sigma) P_{a,s}. \quad (18)$$

The solution of Eqs. (16), (17a), and (17b) is periodic and the period is

$$T = 8K(k)/g(R^2 + a)^{1/2},$$

where $K(k)$ is the complete elliptic integral of the first kind.

We shall now consider in greater detail the solution with the initial conditions corresponding to the absence of interatomic correlation at the initial moment. As in Sec. 3, we shall assume that $R = N_0$ (N_0 is the total number of atoms), $P_s(0) = 2N_1(0)$, and $P_a(0) = 2N_2(0)$. The last two conditions correspond to the initial scattering intensities due to the noncoherent Raman effect. If $N_1(0) \sim N_2(0) \sim N_0$, we have $a \approx 16N_0 \ll N_0^2$ and $k^2 \approx 1 - 32/N_0$. If $k \sim 1$, then in the half-period $[T/4, 3T/4]$, we find that

$$\operatorname{sn}(u, k) \sim -\operatorname{th} u, \quad \frac{P_s}{P_a} \sim \frac{1}{N_0}, \quad P_s \sim \frac{N_0^2}{4} \operatorname{sech}^2 u,$$

and in the next half-period $[3T/4, 5T/4]$, we obtain

$$\operatorname{sn}(u, k) \sim \operatorname{th} u, \quad \frac{P_s}{P_a} \sim \frac{1}{N_0}, \quad P_s \sim \frac{N_0^2}{4} \operatorname{sech}^2 u.$$

Since $K(k) \sim \frac{1}{2} \ln N_0/2$ when $k \sim 1$, it follows that

$$T \sim \frac{4}{gN_0} \ln \frac{N_0}{2} = 4t_0,$$

where t_0 is the delay time of the scattering pulse in the resonance case.

Thus, in the absence of the initial moment of interatomic correlation, the Raman scattering is periodic

with successive emission of the Stokes and anti-Stokes components, and the pulse shape of each component in half a period is similar to the shape of the resonance Raman scattering pulses.

5. DISCUSSION

We shall now consider whether the effect described above can be observed experimentally. The effect appears only if the growth time of a pulse t_0 is shorter than the atomic dephasing time T_2 . It is physically clear that, as in the spontaneous cooperative emission from atoms (see^[3]), the logarithm in Eq. (9) for t_0 should not include the total number of atoms N_0 but μN_0 , where $\mu = 3\lambda^2/4\pi r^2 \ll 1$. This factor is related to the circumstance that the intensity of the emission of a set of N_0 atoms in phase occupying a volume $V = \pi r^2 l$ is proportional to μN_0^2 . The absence of such a factor from Eq. (9) is the result of the one-dimensional approximation.

We shall now substitute in the expression for t_0 the values given in^[6], where the scattering of ruby laser radiation by the $4P_{3/2} - 6S_{1/2} - 5P_{3/2}$ atomic transition in potassium vapor was observed:

$$\gamma = 0.64 \cdot 10^7 \text{ sec}, \quad |G_{1n}/\Omega_{1n}|^2 \sim 0.1, \quad N_0/V = 10^{15} \text{ cm}^{-3}, \\ \lambda = 3.67 \mu, \quad T_2 = 0.5 \cdot 10^{-9} \text{ sec}, \quad l = l_{\max} = c\tau_c$$

Then, $t_0 \sim 10^{-11}$ sec, i.e., the condition $t_0 < T_2$ is satisfied. The characteristic spectral width of the scattered radiation measured in^[6] agrees in order of magnitude with τ_c^{-1} . However, the absence of any special measurements of the pulse shape does not allow us to draw final conclusions.

We can obtain an estimate for the scattering by vibrational levels of molecules in a condensed phase by expressing t_0 in terms of the Raman scattering cross section Q :

$$t_0 = \left[Q \frac{3}{8} \frac{c^2 N l}{\omega^3 \hbar V} |E(k_i)|^2 \right]^{-1} \ln \mu N. \quad (19)$$

Substituting into Eq. (19) the values $Q = 10^{-28} \text{ cm}^2$, $\omega = 10^{15} \text{ sec}^{-1}$, $N/V = 10^{22} \text{ cm}^{-3}$, and $l_{\max} = c\tau_c$, we find that for $T_2 = 5 \times 10^{-12}$ sec the inequality $t_0 \lesssim T_2$ is satisfied when the power density is $P \gtrsim 10^3 \text{ MW/cm}^2$, which is a realistic value.

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