# THEORY OF RADIATION FROM A SYSTEM OF WEAKLY INTERACTING PARTICLES

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Submitted January 28, 1972

Zh. Eksp. Teor. Fiz. 63, 813-819 (September, 1972)

The Hamiltonian for the interaction between an external electromagnetic field and an atomic system is obtained by taking into account the retarded part of the Coulomb interaction between them. The intensity of emission (absorption) of the electromagnetic radiation is calculated with aid of the Hamiltonian. Allowance for the retarded part of the Coulomb interaction between atoms results in the appearance of new (as well as the usual) terms in the formulas for the intensity. Numerical estimates show that allowance for the new terms is particularly important in the infrared frequency range, providing the exciting field is coherent. As an example, the intensity of supermissive signals contains, beside the usual term  $\sim N^2$ , also a number of terms of higher powers of N (N is the number of atoms). The Hamiltonian is used to calculate the intensity of superemissive signals of the light induction or echo type. An analysis of the derived expressions shows that along with emission at the fundamental frequency  $\omega_{12}$ , a system of atoms may emit (absorb) under certain conditions detectable power at the double frequency  $2\omega_{12}$  ( $\omega_{12}$  is the splitting frequency in the spectrum of an isolated atom). For N = 2 this corresponds to a simultaneous radiative transition of both atoms to the ground (excited) state.

## 1. INTRODUCTION

I N the investigation of various processes connected with the study of the interaction of radiation with matter, it is customary to use a model in which the substance is regarded as an aggregate of a large number of quantummechanical particles (atoms, molecules) that interact weakly with one another. The concept of weak interaction means in this case that the energy states of the individual particles are practically independent of the environment, so that the unperturbed Hamiltonian  $\hat{\mathcal{H}}_0$  of the substance is a sum of Hamiltonians  $\hat{\mathcal{H}}_{01}$  that are completely determined by the internal parameters of the particles in question.

In the construction of the interaction Hamiltonian  $\widehat{\mathscr{H}}_1$ of the system under consideration in an external field, it is assumed that each atom interacts with the field independently of the other atoms<sup>[1]</sup>. Then  $\hat{\mathscr{H}}_1$  is likewise a sum of Hamiltonians  $\hat{\mathscr{H}}_{\mathbf{i}\mathbf{i}}$  describing the interaction of the i-th atom with the external field. Such a representation describes well the processes of interaction of a system of atoms with incoherent fields. In this case, the intensity of the occurring processes is equal to  $I_0 N,\,$ where  $I_0$  is the intensity of the corresponding process for the individual atom and N is the number of particles. However, in the study of the processes of emission (absorption) of coherent fields, as will be shown below, the contribution made to the intensity of the processes under consideration by the retarded part of the Coulomb interaction between the atoms becomes significant. Thus, in superradiative signals of the optical induction and echo type [2,3] there appear in addition to the ordinary intensity component proportional to N<sup>2</sup>, also additional terms proportional to  $N^3$  and  $N^4$ , the magnitudes of which can be comparable with the component proportional to  $N^2$ . This means that the concentration dependence of the intensity of the type of optical induction and echo signals differs from the dependence originally

proposed. The allowance for the retarded part of the Coulomb interaction can be described in illustrative fashion by a simple physical model.

The alternating field  $\mathbf{E}_{i}$  acting on the i-th atom of the system constitutes the sum of the external field  $\mathbf{E}_{oi}$ and of the internal field due to the joint action, on the i-th atom, by the moving dipoles of the system, which in turn are situated in the external field. Then  $E_i = E_{oi}(1 + \kappa)$ , where  $\kappa \sim N$  is determined by the polarization properties of the system. If the external alternating field is incoherent, then  $\kappa = 0$ . Since the intensity  $I_0$  of the emission (absorption) by a single atom is proportional to the square of the amplitude of the intensity of the field acting on it, it follows that  $I_0 \sim E_{0i}^2 (1 + \kappa)^2$ . Then the intensity of the superradiative signals of the induction and echo type, which is proportional to  $I_0 N^2$ , will become proportional to  $N^2 (1 + \kappa)^2$ , and this explains the appearance in the intensity of terms proportional to N<sup>3</sup> and N<sup>4</sup>. In addition, the same retarded part of the Coulomb interaction leads to the appearance in the emission spectrum of components whose frequencies are equal to double the frequencies in the emission spectrum of the isolated atom. And although the intensities of these components are much lower than those of the fundamental ones, they may turn out to be appreciable in all kinds of nonlinear processes.

### 2. HAMILTONIAN OF INTERACTION OF A SYSTEM OF WEAKLY INTERACTING ATOMS WITH AN EXTERNAL FIELD

In classical field theory, the Hamiltonian of a system of moving charges is given, accurate to  $v^2/c^2$  (v is the charge velocity and c is the velocity of light, v < c) by<sup>[4]</sup>

$$\begin{aligned} & \mathcal{H} = \sum_{i} \frac{p_i^z}{2m_i} + \sum_{i>j} \frac{e_i e_j}{r_{ij}} - \sum_{i} \frac{p_i^4}{8c^2 m_i^3} \\ & - \sum_{i>j} \frac{e_i e_j}{2c^2 m_i m_j r_{ij}} [\mathbf{p}_i \mathbf{p}_j + (\mathbf{p}_i \mathbf{n}_{ij}) (\mathbf{p}_j \mathbf{n}_{ij})], \end{aligned}$$

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$$\mathbf{n}_{ij} = \mathbf{r}_{ij} / r_{ij}, \tag{2.1}$$

where e<sub>i</sub>, m<sub>i</sub>, and **p**<sub>i</sub> are respectively the charge, mass, and momentum of the i-th charge, and r<sub>ii</sub> is the distance between the charges i and j. Since the velocity of the nuclei is much lower than the velocity of the electrons, the momenta of the nuclei in (2.1) can be set equal to zero. The Hamiltonian of a system of moving charges in an external electromagnetic field defined by the potentials **A** and  $A_0$  is obtained from (2.1) by the transformation  $\mathbf{p}_i \rightarrow \mathbf{p}_i - \mathbf{e}_i \mathbf{A}_i / \mathbf{c}$  we can choose  $\mathbf{A}_0 = 0$  for electromagnetic waves). The subsequent transition to the quantum-mechanical Hamiltonian is effected in accordance with the correspondence principle by replacing the physical quantities with the corresponding operators (see, for example, [5]). Part of the obtained Hamiltonian, including the operators  $\hat{A_i}$ , is the interaction operator of the quantum-mechanical system with the external alternating field.

Expanding the vector potential  $\hat{A}_i$  in plane waves<sup>[1,4]</sup> and confining ourselves to single-photon processes, i.e., retaining in the interaction Hamiltonian only terms linear in the field operators, we write down the interaction Hamiltonian in the form of the sum  $\hat{\mathscr{X}}_1 + \hat{\mathscr{X}}_2$ , where

$$\hat{\mathscr{H}}_{i} = -\frac{e}{m} \sqrt{\frac{2\pi\hbar}{V\omega_{k}}} \sum_{j,k,i} \hat{\mathbf{p}}_{i} \mathbf{e}_{jk} [\hat{a}_{jk} e^{-i\mathbf{k}\mathbf{r}_{i}} + \hat{a}_{jk} e^{i\mathbf{k}\mathbf{r}_{i}}], \qquad (2.2)$$

$$\hat{\mathscr{H}}_{2} = \frac{e^{3}}{2c^{2}m^{2}} \sqrt{\frac{2\pi\hbar}{V\omega_{k}}} \sum_{j,\mathbf{k},i\neq j} \frac{1}{r_{ij}} \left[\hat{\mathbf{p}}_{i}\mathbf{e}_{fk} + (\hat{\mathbf{p}}_{i}\mathbf{n}_{ij})(\mathbf{e}_{fk}\mathbf{n}_{ij})\right] \\ \times \left[\hat{a}_{jk}e^{-i\mathbf{k}\mathbf{r}_{j}} + \hat{a}_{jk}^{*}e^{i\mathbf{k}\mathbf{r}_{j}}\right], \quad \hat{a}_{jk} \sim e^{-i\omega_{k}t},$$
(2.3)

where  $\hat{a}_{fk}^*(\hat{a}_{fk})$  are the creation (annihilation) operators of a photon with wave vector k and polarization f (f = 1 or 2) in the representation of the occupation numbers n of the photon states,  $\mathbf{r}_i$  is the radius vector of the i-th atom, V is the volume of the system of atoms, and  $\omega_k$ is the frequency. Consequently, besides the customarily employed interaction Hamiltonian  $\hat{\mathcal{H}}_1$ , allowance for the retarded part of the Coulomb interaction between the atoms leads to the appearance of an additional interaction Hamiltonian  $\hat{\mathcal{H}}_2$ , which represents the Hamiltonian of the interaction of the system of atoms with the internal alternating field.

### 3. INTENSITY OF EMISSION AND ABSORPTION OF AN ALTERNATING FIELD BY A SYSTEM OF WEAKLY-INTERACTING ATOMS IN THE LONG-WAVE APPROXIMATION

The matrix element of the transition of a system from a state  $\langle a |$  into a state  $\langle b |$  under the influence of a perturbation  $\mathcal{H}_1 + \mathcal{H}_2$ , followed by emission of a photon with polarization  $\mathbf{e_{fk}}$ , frequency  $\omega_{\mathbf{k}}$ , and wave vector  $\mathbf{k}$ , is given by (we assumed the nuclei to be secured at the points  $\mathbf{a}_i$ ):

$$+ \frac{c}{2c^2m^2} K \langle b | \sum_{i+j} \frac{1}{a_{ij}} e^{-i\mathbf{k}\mathbf{a}_j} [\hat{\mathbf{p}}_i \mathbf{e}_{f\mathbf{k}} + (\hat{\mathbf{p}}_i \mathbf{n}_{ij}) (\mathbf{e}_{f\mathbf{k}} \mathbf{n}_{ij})] e^{-i\mathbf{k}\mathbf{b}_j} |a\rangle,$$
  
where

$$K \equiv \sqrt{2\pi\hbar(n+1)} / V\omega_{\mathbf{k}},$$

 $\mathbf{r}_{i} = \mathbf{a}_{i} + \boldsymbol{\xi}_{i}$ , and  $\boldsymbol{\xi}_{i}$  is the radius-vector of the i-th electron relative to its nucleus. In the long-wave approximation<sup>[6]</sup> we can expand  $\exp(i\mathbf{k}\cdot\boldsymbol{\xi}_{i})$  in a series in  $k\boldsymbol{\xi}$ 

 $\ll$  1. We confine ourselves to the first term of the expansion in the dipole approximation. Then, taking into account the transformation

$$\langle 1 | \hat{\mathbf{p}}_i | 2 \rangle = i e^{-i} m \omega_{12} \langle 1 | \hat{\mathbf{d}}_i | 2 \rangle,$$

where  $\omega_{12} = \hbar^{-1}(E_1 - E_2)$ ,  $E_1$  and  $E_2$  are the energies of the states  $\langle 1 |$  and  $\langle 2 |$  of the atom, and  $d_i = e\xi_i$  is the operator of the i-th dipole moment, the matrix element takes the form

$$\mathcal{M}_{a,\bar{n}}^{b,n+1} = -i\omega_{12}K\left\langle b\right| \sum_{i} e^{-i\mathbf{k}\mathbf{a}_{i}} \left(\mathbf{d}_{i}\mathbf{e}_{f\mathbf{k}}\right) \left| a\right\rangle + \frac{ie^{2}\omega_{12}}{2c^{2}m}K$$

$$\times \left\langle b\right| \sum_{i\neq j} \frac{1}{a_{ij}} e^{-i\mathbf{k}\mathbf{a}_{f}} \left[\mathbf{d}_{i}\mathbf{e}_{f\mathbf{k}} + \left(\mathbf{d}_{i}\mathbf{n}_{ij}\right) \left(\mathbf{e}_{f\mathbf{k}}\mathbf{n}_{ij}\right)\right] \left| a\right\rangle.$$
(3.2)

Let us consider a concrete situation in which each atom has an essentially non-equidistant spectrum. Then, in the case of an interaction with an alternating field whose frequency resonates with one of the transitions in the spectrum of the isolated atom, we can carry out the subsequent analysis in terms of a two-level system<sup>[7,8]</sup>. In this case, any physical quantity pertaining to the isolated atom is described by a second-order matrix that can be expanded in four linearly-independent matrices. It is customary to use for this purpose Pauli matrices and the unit matrix<sup>[7,8]</sup>. In this representation, the diploe moment of the atom takes the form

$$\mathbf{d}_{i} = \sqrt{2}d_{0}(\mathbf{x}_{0}\hat{\sigma}_{\mathbf{x}i} + \mathbf{y}_{0}\hat{\sigma}_{\mathbf{y}i}), \qquad (3.3)$$

where  $d_0$  is the absolute value of the matrix element of the operator of the electric dipole moment of the individual atom between the states  $\langle 1|$  and  $\langle 2|$ ,  $x_0$  and  $y_0$  are unit vectors along the coordinate axis x and y, and  $\hat{\sigma}_{xi}$  and  $\hat{\sigma}_{yi}$  are Pauli matrices. In this representation, the eigenfunctions describing the states of the system of atoms are the eigenfunctions of the operators  $\hat{R}^2$  and  $\hat{R}_{\sigma}$ , where

$$\hat{R}^2 = \hat{R}_{x}^{\ 2} + \hat{R}_{y}^{\ 2} + \hat{R}_{z}^{\ 2}, \quad \hat{R}_{a} = \frac{1}{2} \sum_{i} \hat{\sigma}_{ai}.$$

Substituting (3.3) in (3.2) and introducing

$$\hat{R}_{\pm,\mathbf{k}}^{(i)} = \sum_{i \neq j} \hat{R}_{\pm i} \frac{1}{a_{ij}} e^{\pm i\mathbf{k}\mathbf{a}_j} \left[ \left( e_{j,\mathbf{k}}^{(\mathbf{x})} \mp i e_{j,\mathbf{k}}^{(\mathbf{y})} \right) + \left( n_{ij}^{(\mathbf{x})} \mp i n_{ij}^{(\mathbf{y})} \right) \left( e_{j,\mathbf{k}} \mathbf{n}_{ij} \right) \right],$$

$$\hat{R}_{\pm \mathbf{k}} = \sum_{i} e^{\pm i\mathbf{k}\mathbf{a}_i} \hat{R}_{\pm i} \left( e_{j,\mathbf{k}}^{(\mathbf{x})} \mp i e_{j,\mathbf{k}}^{(\mathbf{y})} \right),$$

$$e_{i,\mathbf{k}}^{(\mathbf{x})} = e_{i,\mathbf{k}} \mathbf{x}_0, \quad e_{i,\mathbf{k}}^{(\mathbf{y})} = e_{i,\mathbf{k}} \mathbf{y}_0,$$

we obtain an expression for the matrix element of the transition between the states  $RM \rightarrow RM - 1$  (R is the cooperation number and M is its projection on the quantization axis):

$$\mathcal{M}_{B,M,n}^{R,M-1,n+1} = -i\omega_{12}\sqrt{2} \, d_0 K \langle R M - 1 | \hat{R}_{-k} | R M \rangle (e_{jk}^{(x)} + ie_{jk}^{(y)}) + \frac{i\sqrt{2} \, d_0 e^2 \omega_{12}}{2c^2 m} K \langle R M - 1 | \hat{R}_{-k}^{(1)} | R M \rangle.$$
(3.4)

The intensity at which a system of atoms emits quanta of frequency  $\omega_{\mathbf{k}}$ , polarization  $e_{\mathbf{fk}}$ , and wave vector in the interval  $\mathbf{k}$ ,  $\mathbf{k} + d\mathbf{k}$  is given by

$$dI_{f} = 2\pi\omega_{\mathbf{k}}\mathcal{M}_{R,M-1,n+1}^{R,M,n}\mathcal{M}_{R,M,n}^{R,M-1,n+1}\delta(E_{2}-E_{1}+\hbar\omega_{\mathbf{k}})\frac{Vd\mathbf{k}}{(2\pi)^{3}}.$$
 (3.5)

Following<sup>L7]</sup>, it is easy to show that for mixed states described by a density matrix  $\hat{\rho}(t)$  at an arbitrary instant of time t, the intensity of the spontaneous emission (3.5) into a solid-angle element d $\Omega$  is

$$I_{f}^{(D)}(t) = I_{0f}^{(D)} [\operatorname{Sp}(\hat{\rho}(t) \hat{R}_{+k} \hat{R}_{-k}) + \frac{e^{4}}{4c^{4}m^{3}} \operatorname{Sp}(\hat{\rho}(t) \hat{R}_{+k}^{(1)} \hat{R}_{-k}^{(1)}) - \frac{!e^{3}}{2c^{2}m} \operatorname{Sp}(\hat{\rho}(t) \hat{R}_{+k}^{(1)} \hat{R}_{-k}) - \frac{e^{2}}{2c^{2}m} \operatorname{Sp}(\hat{\rho}(t) \hat{R}_{+k} \hat{R}_{-k}^{(1)})], \qquad (3.6)$$

where  $I_{of}^{(D)}$  is the intensity of emission by a single atom into a solid-angle element in the direction k, and takes the form

$$I_{of}^{D} = \frac{\omega_{12}}{\pi c^3} d_0^2 d\Omega.$$
 (3.7)

The first term of (3.6) is the expression obtained by Dicke<sup> $\lceil 7 \rceil$ </sup> for the emission intensity.

#### 4. INTENSITY OF QUADRUPOLE-TYPE EMISSION FROM A SYSTEM OF TWO-LEVEL ATOMS

Usually inclusion of the second term of the expansion of  $exp(i\mathbf{k} \cdot \boldsymbol{\xi}_i)$  leads to the appearance of electric quadrupole and magnetic dipole transitions between states of an individual atom. When the retarded part of the Coulomb interaction is taken into account, principally new terms of the type  $\mathbf{R}_{\pm i}\mathbf{R}_{\pm i}$  appear in the interaction Hamiltonian. These terms describe a simultaneous dipole transition of two atoms with emission or absorption of a single field quantum of double frequency  $2\omega_{12}$ . Such transitions correspond to a quantum-number change  $\triangle M = \pm 2$ , and will be called quadrupole type transitions, in analogy with the quadrupole transitions with change of the projection of the angular momentum in the spectrum of the individual atom. Let us calculate the intensity of such a process. To this end we retain in the matrix element (3.1) the expansion term  $i\mathbf{k} \cdot \boldsymbol{\xi}_i$ ; changing over to the effective-spin formalism, we then obtain

$$I_{f}^{(q)}(\mathbf{k},t) = I_{0f}^{(q)} \operatorname{Sp}[\hat{\rho}(t) (\hat{R}_{+\mathbf{k}} \hat{R}_{+})^{(1)} (\hat{R}_{-\mathbf{k}} \hat{R}_{-})^{(1)}], \qquad (4.1)$$

$$(\hat{R}_{\pm k}\hat{R}_{\pm})^{(1)} = \sum_{i \neq j} \frac{1}{a_{ij}} e^{\pm i\mathbf{k}\mathbf{a}_j} [(e_{jk}^{(\mathbf{x})} \mp i e_{jk}^{(\mathbf{y})}) + (n_{ij}^{(\mathbf{x})} \mp i n_{ij}^{(\mathbf{y})}) (e_{jk}\mathbf{n}_{ij})] \hat{R}_{\pm i}\hat{R}_{\pm j}.$$
(4.2)

The quantity  $I_{of}^{(Q)}$  is given by the following formula:

$$I_{of}^{(Q)} = \frac{4}{\pi} (n+1) \frac{\omega_{12}{}^{5} d_{0}{}^{4} e^{2}}{c^{8} m^{2}} d\Omega, \qquad (4.3)$$

where n are the occupation numbers for quanta of frequency  $2\omega_{12}$ , polarization  $\mathbf{e}_{\mathbf{fk}}$ , and wave vector  $\mathbf{k}$ .

#### 5. DISCUSSION OF RESULTS

The density matrix of a system of weakly interacting atoms in the state of thermodynamic equilibrium constitute a product of a density matrices  $\hat{\rho}_{oi}$  describing the behavior of isolated atoms. In the two-level approximation, each density matrix  $\hat{\rho}_{01}$  is diagonal and of second order. The intensity of the considered processes of emission (absorption) by such a system of atoms is therefore determined by the diagonal parts of the intensity operators in (3.6) and (4.1). For a two-level system, the only such operators are  $\hat{R}_{\pm i}\hat{R}_{\mp i}$ , which pertain to one and the same isolated i-th particle. Estimates show that the most significant part of the thermal radiation is described by the corresponding part of the first term in (3.6). A similar situation remains also in the case of incoherent excitation of the system. Under coherent excitation on the other hand, off-diagonal matrix

elements appear in the density matrix of each individual atom<sup>[10]</sup>. The matrix elements  $\hat{R}_{\pm i}\hat{R}_{\mp j}$  then make a contribution to the emission (absorption) intensity.

Assume that the system is acted upon by a coherent electromagnetic field in the form (see, for example,  $\lfloor 6 \rfloor$ )

$$\mathbf{A}_{j} = \frac{i}{2} A \mathbf{e}_{j \mathbf{k}_{0}} \mathbf{e}^{-i\mathbf{k}_{0}\mathbf{r}}_{j} e^{i\omega t} + \frac{i}{2} A \mathbf{e}_{j \mathbf{k}_{0}} e^{i\mathbf{k}_{0}\mathbf{r}}_{j} e^{-i\omega t}, \qquad (5.1)$$

where  $\mathbf{e}_{\mathbf{fk}_0}$  is a unit vector determining the emission polarization (the direction of the electric vector),  $\mathbf{k}_0$  is the wave vector, and A is the amplitude of the vector potential. Let us calculate the emission intensity corresponding to the free-induction signal in accordance with formula (3.6). After simple transformations we obtain

$$I_{t}^{(D)}(\mathbf{k},t) = I_{tt}^{(D)}(\mathbf{k},t) \left(1 + A_{1} + A_{2}\right);$$

$$A_{1} = \frac{e^{4}}{e^{4}} \frac{|B|^{2}}{|C|^{4}}, \quad A_{2} = -\frac{e^{2}}{2} \left(\frac{B}{C} + \frac{B^{*}}{C^{*}}\right);$$
(5.2)

$$B = \sum_{\substack{i \neq j \\ i \neq j}} a_{ij}^{-1} e^{+i\mathbf{k}_{B_{j}}} [(e_{jk}^{(s)} - ie_{jk}^{(s)})]$$
(5.3)

$$(n_{ij}^{(\mathbf{z})} - in_{ij}^{(\mathbf{y})}) (\mathbf{e}_{l\mathbf{k}}\mathbf{n}_{ij}) ]e^{-i\mathbf{k}_{\mathbf{y}}\mathbf{a}_{l}} (e_{l\mathbf{k}}^{(\mathbf{z})} + ie_{l\mathbf{k}}^{(\mathbf{y})});$$

$$C = \sum e^{i(\mathbf{k}-\mathbf{k}_{\mathbf{y}})\mathbf{a}_{l}}$$
(5.4)
$$(5.5)$$

$$\int_{a} e^{i(\mathbf{k}-\mathbf{k}_{0})\mathbf{a}_{j}}$$
(5.5)

where  $I_{if}^{(D)}(\mathbf{k}, t) = I_{of}^{(D)} \operatorname{Sp} [\hat{\rho}(t) \hat{R}_{+\mathbf{k}} \hat{R}_{-\mathbf{k}}]$  in the absence of  $\mathcal{H}_2$  coincides with the formula obtained by Dicke for the spontaneous-emission intensity. To estimate the relative contributions of the second and third terms in (5.2)it is necessary to calculate the sums entering in (5.4) and (5.5). We shall calculate the sums B and C for a rigid cubic lattice. The contribution made to B by the second term in the square brackets is always smaller than that of the first, and will be neglected in the subsequent calculations. After substituting the values of the lattice sums [11,3] and integrating over the directions of the wave vector  $\mathbf{k}$ , the formula for the intensity (5.2) becomes

$$I^{(D)}(t) = I_{i}^{(D)}(t) (1 + \alpha N^{2} + \beta N), \qquad (5.6)$$

$$\alpha = \frac{e^{4}}{4c^{4}m^{2}} \left(\frac{4\pi}{V}\right)^{2} \frac{1}{|\mathbf{k}|^{4}}, \quad \beta = -\frac{4\pi e^{2}}{Vc^{2}m|\mathbf{k}|^{2}}.$$
 (5.7)

It follows from (5.6) and (5.7) that (i) the concentration dependence of the intensity of the induction signal differs from that obtained from the previously known formulas, and (ii) the angular distribution of the intensity of coherent radiation coincides in the present approximation with the angular distribution of the intensity obtained without allowance for  $\mathcal{H}_2$ . In the infrared region ( $\omega \sim 10^3 - 10^4$  Hz), for linear sample dimensions L = 1 cm and N  $\sim 10^{20} - 10^{21}$ , the contribution to the emission intensity from the second and third terms of (5.5) is predominant. With further decrease of frequency (down to the radio frequencies), the contribution for the additional terms in (5.6) becomes even more appreciable. Their influence on the concentration dependence should therefore come into play strongly in experiments aimed at the observation of superradiative signals from electric dipole transitions under the influence of a radio-frequency electric field. Such transitions were observed experimentally in a number of studies [12] in which the following substances were used:  $Ti^{3+}$  ions in corundum,  $Co^{2+}$  ions in CdTe,  $Pr^{3+}$  ions in

 $La_{1.98}Pr_{0.02} - Zn(NO_3)24H_2O$ ,  $Cr^{3^+}$  ions in LiNbO<sub>3</sub>, and finally  $Co^{3^+}$  ions in YGaG. The concentration dependence of superradiative transitions can also be investigated in gases, for example in SF<sub>6</sub> gas using a  $CO_2$  laser<sup>[13]</sup>.

An estimate of the emission intensity at the double frequency  $2\omega_{12}$  given by formula (4.1) shows readily that the ratio of the intensity of quadrupole-type emission to the emission intensity (3.6) is proportional to  $(kd_0/e)^2$ . Although this ratio is small, formula (4.1) describes in principle a new type of radiation in all of the frequency bands. In the case of nonlinear processes in samples whose dimensions are much larger than the incident wavelength, the emission can become amplified and lead to generation of a harmonic of comparable power.

The authors are sincerely grateful to the participants of the seminar on solid-state physics, headed by Professor U. Kh. Kopvillem, for a fruitful discussion.

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Translated by J. G. Adashko

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