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## DERIVATION OF A MODEL HAMILTONIAN IN THE “SHORT TIME SCALE” LIMIT

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We investigate the conventional Hamiltonian describing the non-relativistic quantum electrodynamics and the dynamics of the intrinsic states of  $N$  two-level atoms (molecules). It is shown that the Hamiltonian, canonically transformed from the conventional one and including only field operators at the initial time moment, does not contain the near fields inversely proportional to the first and second powers of the distance between any pair of atoms (molecules) on the quite short time scale and allows certain collective radiative effects including the radiation trapping, where atoms or molecules act as a whole.

### 1. Introduction

Today, in a common scientific practice, the non-relativistic quantum theory of radiation is a quite developed and verified experimentally tool of investigation. Many interesting and new phenomena in the domains of superradiance, photon trapping, coherent spectral line broadening, probe beam gain in a strongly pumped medium were in order to be explained during the last several decades. The development of the modern femtosecond light-pulse experimental techniques requires the appropriate revising of the previously built theoretical approaches (like in [1]) based on the assumption that the evolutionary time scale be much bigger than the time of light travel through an atomic system.

The way to describe the possible effects is based on the second quantization of the electromagnetic field coupled with  $N$  quite slowly moving identical atoms or molecules. The methods and the results presented here differ considerably from those of the earlier investigations. In partic-

ular, instead of the construction of evolution equations for a combination of the particle creation and annihilation operators in the Heisenberg representation, we analyze the conventional Hamiltonian (the analysis of the quantization for an electromagnetic field and the applicable restrictions can be found in [2]) for the system and, after its canonical transformation eliminating the time dependence of the field creation and annihilation operators, find the certain limits of the transformed Hamiltonian. In this paper, we generalize the formalism of works [1], [3], and [4] to a spatially degenerated case of non-zero atomic (or molecular) and off-diagonal dipole matrix elements. We consider the non-zero atomic or molecular dipoles in the ground and excited states, all binary combinations of atomic creation and annihilation operators (including such as  $\sigma_i^+ \sigma_j^+$  and  $\sigma_i \sigma_j$ , with  $i \neq j$  – the notations are provided at the beginning of the next section), and short time intervals of the evolution of the atomic (molecular) operators. These intervals are comparable by the order of magnitude with the period of a resonant electromagnetic wave (in the optical region it can reach a few femtoseconds for atomic or molecular transitions to an electronic excited state).

As compared with works [1] and [3], where well-localized atoms were investigated, we explicitly consider the interaction of atoms or molecules with a quantum radiation bath and show that the obtained Hamiltonian including only field operators at the initial time moment does not contain the near fields inversely proportional to the first and second powers of the distance between any pair of atoms (molecules). This Hamiltonian involves also certain radiative effects, including the radia-

tion trapping, where the atoms or molecules act collectively.

## 2. Theory

We discuss here the gas system of  $N$  atoms or molecules interacting with a quantized multimode electromagnetic field. We consider only the atomic (molecular) transitions between two levels  $b$  (corresponding to the ground state) and  $a$  (corresponding to the excited state) and neglect the influence of the translational motion of particles on the absorption (emission) of electromagnetic field quanta during some characteristic time. In this problem, it is the period of electromagnetic waves corresponding to the resonant atomic transition. Then, in the dipole approximation, we describe the dynamics of the  $N$ -atomic intrinsic states along with the state of the electromagnetic field with the help of the following conventional Hamiltonian (the analysis of the dipole approximation can be found, e.g., in [5] and [6]) with the use of the second quantization for an electromagnetic field:

$$\hat{H} = \hbar\omega_a \sum_{i=1}^N \sigma_i^+ \sigma_i + \hbar\omega_b \sum_{i=1}^N \sigma_i \sigma_i^+ + \frac{1}{2} \int d\mathbf{r} (\varepsilon_0 |\mathcal{E}(\mathbf{r})|^2 + \mu_0 |\mathcal{H}(\mathbf{r})|^2) - \sum_{i=1}^N (\wp_{ab}^i \sigma_i^+ + \wp_{ba}^i \sigma_i + \wp_{bb}^i \sigma_i \sigma_i^+ + \wp_{aa}^i \sigma_i^+ \sigma_i) \mathcal{E}(\mathbf{r}_i), \quad (1)$$

where  $\sigma_i^+ = |a\rangle\langle b|_i$  and  $\sigma_i = |b\rangle\langle a|_i$  are the creation and annihilation operators of the excited state for the  $i$ -th particle,  $i = 1..N$ ;  $N$  is number of particles;  $a$  and  $b$  denote the excited and ground states of an atom (molecule), respectively;  $\mathbf{r}_i$  is the position of the  $i$ -th atom or molecule (below, we consider just atoms for brevity). The diagonal and off-diagonal dipole matrix elements are defined by terms  $(\wp_{aa}^i)^* = \wp_{aa}^i = \langle a|\hat{\mu}|a\rangle_i$  and  $(\wp_{bb}^i)^* = \wp_{bb}^i = \langle b|\hat{\mu}|b\rangle_i$  and  $(\wp_{ab}^i)^* = \wp_{ba}^i = \langle b|\hat{\mu}|a\rangle_i$ ,  $i = 1..N$ , respectively, where  $\hat{\mu}$  is the dipole operator for a particle.

In the terms of monochromatic transverse plane waves in the Heisenberg representation, the electric and magnetic field operators  $\mathcal{E}(t, \mathbf{r})$  and  $\mathcal{H}(t, \mathbf{r})$  can be written down as

$$\mathcal{E}(t, \mathbf{r}) = \sum_q \hat{e}_q \mathcal{E}_q e^{i\mathbf{k}_q \mathbf{r}} a_q(t) + \text{H.c.}; \quad (2)$$

$$\mathcal{H}(t, \mathbf{r}) = \frac{1}{\mu_0} \sum_q \frac{1}{\omega_q} [\mathbf{k}_q \times \hat{e}_q] \mathcal{E}_q e^{i\mathbf{k}_q \mathbf{r}} a_q(t) + \text{H.c.}; \quad (3)$$

where  $\hat{e}_q$  is the unit polarization vector perpendicular to the direction of a wave vector  $\mathbf{k}_q$  for the  $q$ -th mode;

$$\mathcal{E}_q = \left( \frac{\hbar\omega_q}{2\varepsilon_0 V} \right)^{1/2}, \quad (4)$$

where  $V$  is a volume allowed to be “filled-in” by the electromagnetic field. Introducing the notation  $a_q(t) = a_q$  and  $a_q^+(t) = a_q^+$  in the Heisenberg representation, where  $a_q$  and  $a_q^+$  are the annihilation and creation operators, respectively, for the  $q$ -th mode. The creation  $a_q^+$  and annihilation  $a_q$  operators for the  $q$ -th mode of the electromagnetic field satisfy the Bose commutation relations  $[a_q(t), a_{q'}^+(t)] = \delta_{qq'}$ .

Taking expressions (2) and (3) into account, we have the following Hamiltonian in the Heisenberg representation neglecting the “zero-point” energy:

$$\hat{H} = \hbar\omega_a \sum_{i=1}^N \sigma_i^+ \sigma_i + \hbar\omega_b \sum_{i=1}^N \sigma_i \sigma_i^+ + \hbar \sum_q \omega_q a_q^+ a_q - \sum_{i=1}^N \sum_q \mathcal{E}_q (\mathbf{s}_i^d + \mathbf{s}_i^o) \hat{e}_q (e^{i\mathbf{k}_q \mathbf{r}_i} a_q + e^{-i\mathbf{k}_q \mathbf{r}_i} a_q^+), \quad (5)$$

where

$$\mathbf{s}_i^d = \wp_{bb}^i \sigma_i \sigma_i^+ + \wp_{aa}^i \sigma_i^+ \sigma_i \quad (6)$$

and

$$\mathbf{s}_i^o = \wp_{ab}^i \sigma_i^+ + \wp_{ba}^i \sigma_i. \quad (7)$$

Using the evolution equations for the field operators  $a_q$  and  $a_q^+$  (in the Heisenberg representation)

$$\frac{d}{dt} a_q = \frac{i}{\hbar} [\hat{H}, a_q] = -i\omega_q a_q + \frac{i}{\hbar} \sum_j \mathcal{E}_j (\mathbf{s}_j^d + \mathbf{s}_j^o) \hat{e}_j e^{-i\mathbf{k}_j \mathbf{r}_j}, \quad (8)$$

we derive the equation that has the sense of a canonical transformation as the consequence of the above canonical equation of motion:

$$a_q(t) = a_q(0) e^{-i\omega_q t} + \frac{i}{\hbar} \sum_j \mathcal{E}_j e^{-i\mathbf{k}_j \mathbf{r}_j} \hat{e}_j \times$$

$$\times \int_0^t dt' (\mathbf{s}_j^d(t') + \mathbf{s}_j^o(t')) e^{-i\omega_q(t-t')}. \quad (9)$$

Substituting (9) in Hamiltonian (5), we finally obtain

$$\begin{aligned} \hat{H} = & \hbar\omega_a \sum_{i=1}^N \sigma_i^+ \sigma_i + \hbar\omega_b \sum_{i=1}^N \sigma_i \sigma_i^+ + \hbar \sum_q \omega_q a_q^+ a_q - \\ & - \sum_{i=1}^N \sum_q \mathcal{E}_q \hat{e}_q [\mathbf{s}_i^d + \mathbf{s}_i^o] \times \\ & \times \left( e^{-i(\omega_q t - \mathbf{k}_q \mathbf{r}_i)} a_q(0) + e^{i(\omega_q t - \mathbf{k}_q \mathbf{r}_i)} a_q^+(0) \right) - \\ & - \frac{i}{\hbar} \sum_{i=1}^N \sum_{j=1}^N \sum_q (\mathcal{E}_q)^2 \hat{e}_q [\mathbf{s}_i^d(t) + \mathbf{s}_i^o(t)] \left\{ e^{i\mathbf{k}_q(\mathbf{r}_i - \mathbf{r}_j)} \hat{e}_q \times \right. \\ & \times \int_0^t dt' [\mathbf{s}_j^d(t') + \mathbf{s}_j^o(t')] e^{-i\omega_q(t-t')} - \\ & \left. - e^{-i\mathbf{k}_q(\mathbf{r}_i - \mathbf{r}_j)} \hat{e}_q \int_0^t dt' [\mathbf{s}_j^d(t') + \mathbf{s}_j^o(t')] e^{i\omega_q(t-t')} \right\}. \quad (10) \end{aligned}$$

Next, we search for the limit of continuous modes as  $V \rightarrow \infty$  in the Hamiltonian above. In this limit, we present the sum over the electromagnetic modes  $q$  through the integration as

$$\begin{aligned} & \frac{1}{V} \sum_q \left\{ \hat{e}_q [\mathbf{s}_i^d(t) + \mathbf{s}_i^o(t)] \right\} \left\{ \hat{e}_q [\mathbf{s}_j^d(t') + \mathbf{s}_j^o(t')] \right\} = \\ & = \frac{1}{V} \sum_{\mathbf{k}} \left[ (\mathbf{s}_i^d(t) + \mathbf{s}_i^o(t)) (\mathbf{s}_j^d(t') + \mathbf{s}_j^o(t')) - \right. \\ & \left. - \left\{ \hat{\mathbf{k}} (\mathbf{s}_i^d(t) + \mathbf{s}_i^o(t)) \right\} \left\{ \hat{\mathbf{k}} (\mathbf{s}_j^d(t') + \mathbf{s}_j^o(t')) \right\} \right] \rightarrow \\ & \rightarrow \lim_{\omega_M \rightarrow \infty} \left( \frac{1}{2\pi c} \right)^3 \int_0^{\omega_M} \omega^2 d\omega \int d\hat{\mathbf{k}} \times \\ & \times \sum_{\alpha, \alpha', \beta, \beta'} \left\{ |\varrho_{\alpha \alpha'}^i| |\varrho_{\beta \beta'}^j| \left[ \left( \hat{\varrho}_{\alpha \alpha'}^i \hat{\varrho}_{\beta \beta'}^j \right) - \right. \right. \end{aligned}$$

$$\left. - \left( \hat{\varrho}_{\alpha \alpha'}^i \hat{\mathbf{k}} \right) \left( \hat{\varrho}_{\beta \beta'}^j \hat{\mathbf{k}} \right) \right] |\alpha\rangle \langle \alpha'|_i(t) |\beta\rangle \langle \beta'|_j(t') \}, \quad (11)$$

where  $\alpha, \alpha', \beta,$  and  $\beta'$  denote  $a$  (excited) or  $b$  (ground); the unit vector  $\hat{\mathbf{k}} = \mathbf{k} = \mathbf{k}_q/k_q$  is parallel to the “direction of propagation” of the  $q$ -th mode, and  $d\hat{\mathbf{k}}$  denotes the infinitesimal space angle. Each  $q$ -th mode includes two orthogonal polarization planes described by two unit vectors  $\hat{e}_1 \perp \hat{e}_2 \perp \hat{\mathbf{k}}$ . The maximum frequency  $\omega_M$  is quite large (physically), but supposed to be in the region of the dipole approximation for the electromagnetic field–atom interaction. Therefore, expression (11) in the described limit becomes

$$\begin{aligned} \hat{H} = & \hbar\omega_a \sum_{i=1}^N \sigma_i^+ \sigma_i + \hbar\omega_b \sum_{i=1}^N \sigma_i \sigma_i^+ + \hbar \sum_q \omega_q a_q^+ a_q - \\ & - \sum_{i=1}^N \sum_q \mathcal{E}_q \hat{e}_q [\mathbf{s}_i^d + \mathbf{s}_i^o] \times \\ & \times \left( e^{-i(\omega_q t - \mathbf{k}_q \mathbf{r}_i)} a_q(0) + e^{i(\omega_q t - \mathbf{k}_q \mathbf{r}_i)} a_q^+(0) \right) - \\ & - \frac{i}{2\epsilon_0} \left( \frac{1}{2\pi c} \right)^3 \sum_{i=1}^N \sum_{j=1}^N \lim_{\omega_M \rightarrow \infty} \int_0^t dt' \int_0^{\omega_M} \omega^3 d\omega \int d\hat{\mathbf{k}} \times \\ & \times \left\{ e^{i\mathbf{k}_q(\mathbf{r}_i - \mathbf{r}_j)} \sum_{\alpha, \alpha', \beta, \beta'} \left\{ |\varrho_{\alpha \alpha'}^i| |\varrho_{\beta \beta'}^j| \left[ \left( \hat{\varrho}_{\alpha \alpha'}^i \hat{\varrho}_{\beta \beta'}^j \right) - \right. \right. \right. \\ & \left. \left. - \left( \hat{\varrho}_{\alpha \alpha'}^i \hat{\mathbf{k}} \right) \left( \hat{\varrho}_{\beta \beta'}^j \hat{\mathbf{k}} \right) \right] |\alpha\rangle \langle \alpha'|_i(t) |\beta\rangle \langle \beta'|_j(t') \right\} \times \\ & \times e^{-i\omega_q(t-t')} - e^{-i\mathbf{k}_q(\mathbf{r}_i - \mathbf{r}_j)} \times \\ & \times \sum_{\alpha, \alpha', \beta, \beta'} \left\{ |\varrho_{\alpha \alpha'}^i| |\varrho_{\beta \beta'}^j| \left[ \left( \hat{\varrho}_{\alpha \alpha'}^i \hat{\varrho}_{\beta \beta'}^j \right) - \right. \right. \\ & \left. \left. - \left( \hat{\varrho}_{\alpha \alpha'}^i \hat{\mathbf{k}} \right) \left( \hat{\varrho}_{\beta \beta'}^j \hat{\mathbf{k}} \right) \right] |\alpha\rangle \langle \alpha'|_i(t) |\beta\rangle \langle \beta'|_j(t') \right\} e^{i\omega_q(t-t')}. \quad (12) \end{aligned}$$

As the next step, we calculate the integral over the space angle. For this, it is enough to find the following integral for a item under the sum over the indices  $\alpha, \alpha', \beta,$  and  $\beta'$ .

To simplify the calculation, the coordinate system for the integration is defined by three orthonormal vectors:  $\hat{e}_z$  is parallel to the vector  $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$  ( $OZ$  axis) connecting the  $j$ -th atom with the  $i$ -th one,  $\hat{e}_x$ , and  $\hat{e}_y$ ; dipole matrix element's unit vectors are presented as  $\hat{\varphi}_{\alpha\alpha'}^i = (\hat{\varphi}_{\alpha\alpha'}^i)_{\parallel} + (\hat{\varphi}_{\alpha\alpha'}^i)_{\perp}$ , where the  $(\hat{\varphi}_{\alpha\alpha'}^i)_{\parallel}$  component is parallel to the vector  $\mathbf{r}_{ij}$  (or the axis  $OZ$ ), and  $(\hat{\varphi}_{\alpha\alpha'}^i)_{\perp}$  lies in the plane created by two unit vectors  $\hat{e}_x$  and  $\hat{e}_y$  (both are perpendicular to  $\mathbf{r}_{ij}$ ). Without any loss of generality, we may assume that  $(\hat{\varphi}_{\alpha\alpha'}^i)_{\perp} \cdot \hat{e}_y = 0$  (only  $x$  component is saved for  $\hat{\varphi}_{\alpha\alpha'}^i$ , that is not true for  $\hat{\varphi}_{\beta\beta'}^j$ ) and  $\hat{k} = \cos\varphi \sin\theta \hat{e}_x + \sin\varphi \sin\theta \hat{e}_y + \cos\theta \hat{e}_z$ . We now have

$$\begin{aligned} (\hat{\varphi}_{\alpha\alpha'}^i \hat{\mathbf{k}}) (\hat{\varphi}_{\beta\beta'}^j \hat{\mathbf{k}}) &= \left[ (\hat{\varphi}_{\beta\beta'}^j \hat{\mathbf{e}}_x) \cos^2\varphi + \right. \\ &+ \left. (\hat{\varphi}_{\beta\beta'}^j \hat{\mathbf{e}}_y) \sin\varphi \cos\varphi \right] (\hat{\varphi}_{\alpha\alpha'}^i \hat{\mathbf{e}}_x) \sin^2\theta + \\ &+ (\hat{\varphi}_{\alpha\alpha'}^i \hat{\mathbf{e}}_x) (\hat{\varphi}_{\beta\beta'}^j \hat{\mathbf{e}}_z) \cos\varphi \sin\theta \cos\theta + \\ &+ (\hat{\varphi}_{\alpha\alpha'}^i \hat{\mathbf{e}}_z) (\hat{\varphi}_{\beta\beta'}^j \hat{\mathbf{e}}_z) \cos^2\theta + (\hat{\varphi}_{\alpha\alpha'}^i \hat{\mathbf{e}}_z) \times \\ &\times \left[ (\hat{\varphi}_{\beta\beta'}^j \hat{\mathbf{e}}_x) \cos\varphi + (\hat{\varphi}_{\beta\beta'}^j \hat{\mathbf{e}}_y) \sin\varphi \right] \cos\theta \sin\theta. \end{aligned} \quad (13)$$

Therefore,

$$\begin{aligned} \int d\hat{\mathbf{k}} e^{i\mathbf{k}_q \mathbf{r}_{ij}} \left[ (\hat{\varphi}_{\alpha\alpha'}^i \hat{\varphi}_{\beta\beta'}^j) - (\hat{\varphi}_{\alpha\alpha'}^i \hat{\mathbf{k}}) (\hat{\varphi}_{\beta\beta'}^j \hat{\mathbf{k}}) \right] &= \\ = \pi \int d\theta \sin\theta \left\{ 2 (\hat{\varphi}_{\alpha\alpha'}^i \hat{\varphi}_{\beta\beta'}^j) - (\hat{\varphi}_{\alpha\alpha'}^i)_{\perp} (\hat{\varphi}_{\beta\beta'}^j)_{\perp} + \right. \\ &+ \left. [(\hat{\varphi}_{\alpha\alpha'}^i)_{\perp} (\hat{\varphi}_{\beta\beta'}^j)_{\perp} - \right. \\ &- 2 (\hat{\varphi}_{\alpha\alpha'}^i)_{\parallel} (\hat{\varphi}_{\beta\beta'}^j)_{\parallel}] \cos^2\theta \left. \right\} e^{i\mathbf{k}_q \mathbf{r}_{ij} \cos\theta} = \\ = 4\pi \left\{ \left[ (\hat{\varphi}_{\alpha\alpha'}^i \hat{\varphi}_{\beta\beta'}^j) - (\hat{\varphi}_{\alpha\alpha'}^i)_{\parallel} (\hat{\varphi}_{\beta\beta'}^j)_{\parallel} \right] \frac{\sin(k_q r_{ij})}{k_q r_{ij}} + \right. \\ &+ \left. \left[ (\hat{\varphi}_{\alpha\alpha'}^i \hat{\varphi}_{\beta\beta'}^j) - 3 (\hat{\varphi}_{\alpha\alpha'}^i)_{\parallel} (\hat{\varphi}_{\beta\beta'}^j)_{\parallel} \right] \times \right. \end{aligned}$$

$$\left. \times \left( \frac{\cos(k_q r_{ij})}{(k_q r_{ij})^2} - \frac{\sin(k_q r_{ij})}{(k_q r_{ij})^3} \right) \right\}. \quad (14)$$

Substituting the integral found above in expression (12) with  $k_q = \frac{\omega}{c}$ , we want to find the integral over the frequency. For this, we use the following expressions (calculated in Appendix):

$$\begin{aligned} I_0 \left( \frac{r_{ij}}{c}, t-t' \right) &= \int_0^{\omega_M} d\omega \sin \left( \frac{r_{ij}}{c} \omega \right) e^{-i\omega(t-t')} \rightarrow \\ &\rightarrow -\frac{i\pi}{2} \left\{ \delta \left( t' - \left( t - \frac{r_{ij}}{c} \right) \right) - \delta \left( t' - \left( t + \frac{r_{ij}}{c} \right) \right) \right\}, \end{aligned} \quad (15)$$

$$I_1 \left( \frac{r_{ij}}{c}, t-t' \right) = \int_0^{\omega_M} d\omega \omega \cos \left( \frac{r_{ij}}{c} \omega \right) e^{-i\omega(t-t')} \rightarrow 0, \quad (16)$$

and, finally,

$$\begin{aligned} I_2 \left( \frac{r_{ij}}{c}, t-t' \right) &= \int_0^{\omega_M} d\omega \omega^2 \sin \left( \frac{r_{ij}}{c} \omega \right) e^{-i\omega(t-t')} \rightarrow \\ &\rightarrow \frac{i\pi}{2} \left\{ \frac{\partial^2}{\partial t'^2} \delta \left( t' - \left( t - \frac{r_{ij}}{c} \right) \right) - \frac{\partial^2}{\partial t'^2} \delta \left( t' - \left( t + \frac{r_{ij}}{c} \right) \right) \right\}. \end{aligned} \quad (17)$$

Therefore, by integrating the reviewed item in (12) over the time and taking into account that  $I_0(0, t-t') = I_1(0, t-t') = I_2(0, t-t') = 0$  for  $r_{ij} = 0$ , we obtain the Hamiltonian

$$\begin{aligned} \hat{H} &= \hbar\omega_a \sum_{i=1}^N \sigma_i^+(t) \sigma_i(t) + \\ &+ \hbar\omega_b \sum_{i=1}^N \sigma_i(t) \sigma_i^+(t) + \hbar \sum_q \omega_q a_q^+(t) a_q(t) - \\ &- \sum_{i=1}^N \sum_q \mathcal{E}_q \hat{e}_q [\mathbf{s}_i^d(t) + \mathbf{s}_i^o(t)] \times \\ &\times \left( e^{-i(\omega_q t - \mathbf{k}_q \cdot \mathbf{r}_i)} a_q(0) + e^{i(\omega_q t - \mathbf{k}_q \cdot \mathbf{r}_i)} a_q^+(0) \right) + \end{aligned}$$

$$\begin{aligned}
 & + \frac{1}{4\pi\epsilon_0} \sum_{i,j;i \neq j}^{N,N} \frac{1}{|\mathbf{r}_i(t) - \mathbf{r}_j(t - \frac{r_{ij}}{c})|^3} \times \\
 & \times \sum_{\alpha,\alpha',\beta,\beta'} |\hat{\rho}_{\alpha\alpha'}^i| |\hat{\rho}_{\beta\beta'}^j| \left[ \left( \hat{\rho}_{\alpha\alpha'}^i(t) \cdot \hat{\rho}_{\beta\beta'}^j(t - \frac{r_{ij}}{c}) \right) - \right. \\
 & \left. - 3 \left( \hat{\rho}_{\alpha\alpha'}^i \right)_\parallel(t) \cdot \left( \hat{\rho}_{\beta\beta'}^j \right)_\parallel(t - \frac{r_{ij}}{c}) \right] \times \\
 & \times |\alpha\rangle\langle\alpha'|_i(t) |\beta\rangle\langle\beta'|_j(t - \frac{r_{ij}}{c}). \tag{18}
 \end{aligned}$$

Let us substitute (9) in (18). We obtain the Hamiltonian in terms of the operators of creation and annihilation at the time  $t_0 = 0$ . This yields the following changes: instead of  $\hbar \sum_q \omega_q a_q^+(t) a_q(t)$ , we have

$$\hbar \sum_q \omega_q a_q^+(0) a_q(0); \tag{19}$$

and

$$\begin{aligned}
 & i \sum_{i=1}^N \sum_q a_q^+(0) \omega_q \mathcal{E}_q e^{-i(\mathbf{k}_q \mathbf{r}_i)} \hat{e}_q \times \\
 & \times \int_0^t dt' [\mathbf{s}_i^d(t') + \mathbf{s}_i^o(t')] e^{i\omega_q t'} + H. c. = \\
 & = \sum_{i=1}^N \sum_q \mathcal{E}_q \hat{e}_q [\mathbf{s}_i^d(\bar{t}) + \mathbf{s}_i^o(\bar{t})] \left\{ a_q(0) e^{i(\mathbf{k}_q \mathbf{r}_i)} \times \right. \\
 & \left. \times [e^{-i\omega_q \bar{t}} - 1] + a_q^+(0) e^{-i(\mathbf{k}_q \mathbf{r}_i)} [e^{i\omega_q \bar{t}} - 1] \right\}, \tag{20}
 \end{aligned}$$

where  $\bar{t} \in [0, t]$  is an averaged time corresponding to the integration of a function changing quite slowly with the time, as compared with a periodic function. The Hamiltonian includes also the term

$$\begin{aligned}
 & \frac{1}{i} \frac{1}{2\epsilon_0} \left( \frac{1}{2\pi c} \right)^3 \sum_{i=1}^N \sum_{j=1}^N \int_0^t dt'' \int_0^t dt' \times \\
 & \times \lim_{\omega_M \rightarrow \infty} \int_0^{\omega_M} \omega^3 d\omega \left\{ \left( \frac{\partial}{\partial t'} e^{-i\omega(t''-t')} \right) \int d\hat{\mathbf{k}} e^{i\mathbf{k}_q(\mathbf{r}_i - \mathbf{r}_j)} \times \right.
 \end{aligned}$$

$$\begin{aligned}
 & \times \sum_{\alpha,\alpha',\beta,\beta'} |\hat{\rho}_{\alpha\alpha'}^i| |\hat{\rho}_{\beta\beta'}^j| |\alpha\rangle\langle\alpha'|_i(t) |\beta\rangle\langle\beta'|_j(t'') \times \\
 & \times \left. \left[ \left( \hat{\rho}_{\alpha\alpha'}^i \hat{\rho}_{\beta\beta'}^j \right) - \left( \hat{\rho}_{\alpha\alpha'}^i \hat{\mathbf{k}} \right) \left( \hat{\rho}_{\beta\beta'}^j \hat{\mathbf{k}} \right) \right] \right\}. \tag{21}
 \end{aligned}$$

By introducing the notation

$$\begin{aligned}
 F(t', t'') & = \sum_{i,j;i \neq j}^{N,N} \frac{1}{|\mathbf{r}_i(t') - \mathbf{r}_j(t'')|^3} \times \\
 & \times \sum_{\alpha,\alpha',\beta,\beta'} |\hat{\rho}_{\alpha\alpha'}^i| |\hat{\rho}_{\beta\beta'}^j| \left[ \left( \hat{\rho}_{\alpha\alpha'}^i(t') \hat{\rho}_{\beta\beta'}^j(t'') \right) - \right. \\
 & \left. - 3 \left( \hat{\rho}_{\alpha\alpha'}^i \right)_\parallel(t') \left( \hat{\rho}_{\beta\beta'}^j \right)_\parallel(t'') \right] |\alpha\rangle\langle\alpha'|_i(t') |\beta\rangle\langle\beta'|_j(t''), \tag{22}
 \end{aligned}$$

we can present it, after the integrations over the space angle and the frequency, for sufficiently short intervals  $[0, t]$  as

$$\begin{aligned}
 & \frac{1}{2} \frac{1}{4\pi\epsilon_0} \int_0^t dt'' \int_0^t dt' \left\{ \frac{\partial}{\partial t'} \left[ \delta \left( t' - \left( t'' - \frac{r_{ij}}{c} \right) \right) - \right. \right. \\
 & \left. \left. - \delta \left( t' - \left( t'' + \frac{r_{ij}}{c} \right) \right) \right] \right\} F(t', t'') = \\
 & = -\frac{1}{2} \frac{1}{4\pi\epsilon_0} \int_0^t dt'' \left[ \frac{1}{2} \frac{\partial}{\partial t''} F \left( t'' - \frac{r_{ij}}{c}, t'' \right) - \right. \\
 & \left. - \frac{1}{2} \frac{\partial}{\partial t''} F \left( t'' + \frac{r_{ij}}{c}, t'' \right) \right] = -\frac{1}{2} \frac{1}{4\pi\epsilon_0} \left[ \frac{1}{2} F \left( t - \frac{r_{ij}}{c}, t \right) + \right. \\
 & \left. + \frac{1}{2} F \left( \frac{r_{ij}}{c}, 0 \right) \right] \approx -\frac{1}{2} \frac{1}{4\pi\epsilon_0} F \left( t - \frac{r_{ij}}{c}, t \right). \tag{23}
 \end{aligned}$$

Here, we took into account that the derivatives of the delta-functions will give non-zero integrals only in the integration region  $[0, t; 0, t]$ . Hence, the “outside” terms  $-F(-\frac{r_{ij}}{c}, 0)$  and  $-F(t + \frac{r_{ij}}{c}, t)$  are zeroed. Moreover, the symmetry of the quadratic form (22) of the function

$F(t'', t'')$  was used to obtain the total derivative under the sign of double integral. For the subsequent integration over the plane  $[0, t; 0, t]$ , we can apply the relation

$$\left( \frac{\partial}{\partial y} F(y, t'') \right)_{y=t''} = \frac{1}{2} \frac{\partial}{\partial t''} F(t'', t''). \quad (24)$$

Thus, substituting (23 and (19) into Hamiltonian (18) and taking into account that term (20) cancels the similar term in the Hamiltonian for sufficiently short time intervals  $[0, t]$ , we obtain the following limit of Hamiltonian (11):

$$\begin{aligned} \hat{H} \approx & \hbar\omega_a \sum_{i=1}^N \sigma_i^+(t) \sigma_i(t) + \hbar\omega_b \sum_{i=1}^N \sigma_i(t) \sigma_i^+(t) + \\ & + \hbar \sum_q \omega_q a_q^+(0) a_q(0) - \sum_{i=1}^N \sum_q \mathcal{E}_q \hat{e}_q [\mathbf{s}_i^d(t) + \mathbf{s}_i^o(t)] \times \\ & \times (e^{i\mathbf{k}_q \mathbf{r}_i} a_q(0) + e^{-i\mathbf{k}_q \mathbf{r}_i} a_q^+(0)) + \\ & + \frac{1}{2} \frac{1}{4\pi\epsilon_0} \sum_{i,j; i \neq j}^{N,N} \frac{1}{|\mathbf{r}_i(t) - \mathbf{r}_j(t)|^3} \times \\ & \times \sum_{\alpha, \alpha', \beta, \beta'} |\hat{\rho}_{\alpha\alpha'}^i| |\hat{\rho}_{\beta\beta'}^j| \left[ \left( \hat{\rho}_{\alpha\alpha'}^i(t) \hat{\rho}_{\beta\beta'}^j(t) \right) - \right. \\ & \left. - 3 \left( \hat{\rho}_{\alpha\alpha'}^i \right)_{\parallel}(t) \left( \hat{\rho}_{\beta\beta'}^j \right)_{\parallel}(t) \right] |\alpha\rangle \langle \alpha'|_i(t) |\beta\rangle \langle \beta'|_j(t). \quad (25) \end{aligned}$$

Here, we assumed that the introduced operator  $F(t', t'')$  is not changed significantly on the intervals  $t', t'' \in [t_0, t = t_0 + \Delta t]$  such that  $\Delta t > r_{ij}/c$  for any pair of atoms (molecules)  $i$  and  $j$ :

$$\frac{\partial}{\partial t'} F(t', t'') \Delta t \ll F(t', t''). \quad (26)$$

As a result, while summing over the atomic indices  $i$  and  $j$  in Hamiltonian (25), we have to consider only such pairs of atoms, whose coordinates satisfy the condition  $\Delta t > r_{ij}/c$ . Other pairs with  $r_{ij} > c\Delta t$  will not contribute in accordance with the obtained expression (15) for the integral  $I_0\left(\frac{r_{ij}}{c}, t-t'\right)$  (15).

### 3. Conclusion

In this paper, we have obtained the model Hamiltonian for the  $N$ -atomic (molecular) system in a continuum of quantized electromagnetic modes with regard for the possibility of a two-photon excitation or decay (jumps) involving a pair of atoms interacting with a dipole-dipole coupling. For comparison, the earlier theoretical work [3] described the evolution of two dipole-dipole interacting atoms in a vacuum with only one atom being initially excited; work [7] considered two two-level atoms independently interacting with local thermal or squeezed reservoirs, taking the possibility of their initial simultaneous excitation into account, but neglecting the dipole-dipole interaction; works [8] and [9] follow the approximations of [1] and [4], just adding an additional state corresponding to two simultaneously excited atoms to the model that differs basically from our description.

The obtained Hamiltonian allows to model the dipole-dipole interaction between atoms or molecules, including the atomic (molecular) interaction with the radiation bath, and to build the microscopic kinetic equations for density matrix elements of the system in a straightforward manner. The more cumbersome approach involving the Green function method (as in [10] - [14]) does not allow one to formulate the kinetic equations in terms of the density matrix describing the probability distribution of states of a system.

The Hamiltonian derived in the present paper contains only field operators at the initial time moment and the atomic (molecular) operators in the Heisenberg representation and does not possess the near fields inversely proportional to the first and second powers of the distance between any pair of particles. The described "short time scale" limit of the Hamiltonian has a dependence inversely proportional to the cube of the distance between atomic pairs and clarifies the collective character of certain radiative effects, including the radiation trapping.

### APPENDIX

Here, we show how the introduced integrals (15), (16), and (17) were calculated.

$$\begin{aligned} I_0(\tau, t-t') &= \int_0^{\omega_M} d\omega \sin(\tau\omega) e^{-i\omega(t-t')} = \\ &= \frac{1}{2} \left\{ -i\pi \frac{\sin[\omega_M(\tau - (t-t'))]}{\pi(\tau - (t-t'))} + i\pi \frac{\sin[\omega_M(\tau + (t-t'))]}{\pi(\tau + (t-t'))} \right\} + \\ &+ \frac{1 - \cos[\omega_M(\tau - (t-t'))]}{\tau - (t-t')} + \frac{1 - \cos[\omega_M(\tau + (t-t'))]}{\tau + (t-t')} \Bigg\}; \quad (27) \end{aligned}$$

and

$$\begin{aligned}
 I_1(\tau, t-t') &= \int_0^{\omega_M} d\omega \omega \cos(\tau\omega) e^{-i\omega(t-t')} = \\
 &= \frac{1}{2} \left\{ \pi\omega_M \frac{\sin[\omega_M(\tau-(t-t'))]}{\pi(\tau-(t-t'))} + i \frac{\sin[\omega_M(\tau-(t-t'))]}{(\tau-(t-t'))^2} - \right. \\
 &- \pi\omega_M \frac{1-\cos[\omega_M(\tau-(t-t'))]}{\pi\omega_M(\tau-(t-t'))^2} + \omega_M \frac{\cos[\omega_M(\tau-(t-t'))]}{i(\tau-(t-t'))} + \\
 &+ \pi\omega_M \frac{\sin[\omega_M(\tau+(t-t'))]}{\pi(\tau+(t-t'))} - i \frac{\sin[\omega_M(\tau+(t-t'))]}{(\tau+(t-t'))^2} - \\
 &- \pi\omega_M \frac{1-\cos[\omega_M(\tau+(t-t'))]}{\pi\omega_M(\tau+(t-t'))^2} - \left. \omega_M \frac{\cos[\omega_M(\tau+(t-t'))]}{i(\tau+(t-t'))} \right\}; \tag{28}
 \end{aligned}$$

and the integral

$$\begin{aligned}
 I_2(\tau, t-t') &= \int_0^{\omega_M} d\omega \omega^2 \sin(\tau\omega) e^{-i\omega(t-t')} = \\
 &\frac{1}{2i} \left\{ \omega_M^2 \left[ \frac{e^{i\omega_M(\tau-(t-t'))}}{i(\tau-(t-t'))} + \frac{e^{-i\omega_M(\tau+(t-t'))}}{i(\tau+(t-t'))} \right] - \right. \\
 &- 2\omega_M \left[ \frac{e^{i\omega_M(\tau-(t-t'))}}{(i(\tau-(t-t')))^2} - \frac{e^{-i\omega_M(\tau+(t-t'))}}{(i(\tau+(t-t')))^2} \right] + \\
 &+ 2 \left[ \frac{e^{i\omega_M(\tau-(t-t'))}}{(i(\tau-(t-t')))^3} + \frac{e^{-i\omega_M(\tau+(t-t'))}}{(i(\tau+(t-t')))^3} \right] - \\
 &- 2 \left[ \frac{1}{(i(\tau-(t-t')))^3} + \frac{1}{(i(\tau+(t-t')))^3} \right] \left. \right\}. \tag{29}
 \end{aligned}$$

The last expression can be rewritten in terms corresponding to the delta-functional sequences, by using the formula

$$\begin{aligned}
 \frac{\sin[\omega_M(\tau-(t-t'))]}{(\tau-(t-t'))^3} &= \frac{1}{2} \left\{ \frac{d^2}{dt'^2} \left( \frac{\sin[\omega_M(\tau-(t-t'))]}{\tau-(t-t')} \right) + \right. \\
 &+ \pi\omega_M^2 \frac{\sin[\omega_M(\tau-(t-t'))]}{\pi(\tau-(t-t'))} + 2\omega_M \frac{\cos[\omega_M(\tau-(t-t'))]}{(\tau-(t-t'))^2} \left. \right\}; \tag{30}
 \end{aligned}$$

so that,

$$\begin{aligned}
 I_2(\tau, t-t') &= \int_0^{\omega_M} d\omega \omega^2 \sin(\tau\omega) e^{-i\omega(t-t')} = \\
 &= \frac{1}{2i} \left\{ \pi\omega_M^2 \frac{\sin[\omega_M(\tau-(t-t'))]}{\pi(\tau-(t-t'))} + 2i\omega_M \frac{\sin[\omega_M(\tau-(t-t'))]}{(\tau-(t-t'))^2} - \right. \\
 &- \left[ \pi \frac{d^2}{dt'^2} \left( \frac{\sin[\omega_M(\tau-(t-t'))]}{\pi(\tau-(t-t'))} \right) + \pi\omega_M^2 \frac{\sin[\omega_M(\tau-(t-t'))]}{\pi(\tau-(t-t'))} + \right. \\
 &+ 2\omega_M \frac{\cos[\omega_M(\tau-(t-t'))]}{(\tau-(t-t'))^2} \left. \right] - i\omega_M^2 \frac{\cos[\omega_M(\tau-(t-t'))]}{\tau-(t-t')} + \\
 &+ 2\omega_M \frac{\cos[\omega_M(\tau-(t-t'))]}{(\tau-(t-t'))^2} - \\
 &- 2i\pi\omega_M \left[ \frac{1}{\tau-(t-t')} \frac{1-\cos[\omega_M(\tau-(t-t'))]}{\pi\omega_M(\tau-(t-t'))^2} \right] \left. \right\}
 \end{aligned}$$

$$\begin{aligned}
 &- \pi\omega_M^2 \frac{\sin[\omega_M(\tau+(t-t'))]}{\pi(\tau+(t-t'))} + 2i\omega_M \frac{\sin[\omega_M(\tau+(t-t'))]}{(\tau+(t-t'))^2} + \\
 &+ \left[ \pi \frac{d^2}{dt'^2} \left( \frac{\sin[\omega_M(\tau+(t-t'))]}{\pi(\tau+(t-t'))} \right) + \pi\omega_M^2 \frac{\sin[\omega_M(\tau+(t-t'))]}{\pi(\tau+(t-t'))} + \right. \\
 &+ 2\omega_M \frac{\cos[\omega_M(\tau+(t-t'))]}{(\tau+(t-t'))^2} \left. \right] - i\omega_M^2 \frac{\cos[\omega_M(\tau+(t-t'))]}{\tau+(t-t')} - \\
 &- 2\omega_M \frac{\cos[\omega_M(\tau+(t-t'))]}{(\tau+(t-t'))^2} - \\
 &- 2i\pi\omega_M \left[ \frac{1}{\tau+(t-t')} \frac{1-\cos[\omega_M(\tau+(t-t'))]}{\pi\omega_M(\tau+(t-t'))^2} \right] \left. \right\}. \tag{31}
 \end{aligned}$$

As is now obvious, the terms non-integrable over the time,  $\frac{\cos[\omega_M(\tau \pm (t-t'))]}{(\tau \pm (t-t'))^2}$ , cancel one another. In addition, two delta-functional sequences  $\frac{\sin[\omega_M(\tau \pm (t-t'))]}{\pi(\tau \pm (t-t'))}$ , each having its corresponding pair with opposite sign, cancel each other.

We now are ready to complete the calculation of the integral over the time in Hamiltonian (12). We present the delta-functional sequences under the time integral in the “large” (physically) frequency limit (formally  $\omega_M \rightarrow \infty$ ) through the corresponding delta-functions:

$$\frac{\sin[\omega_M(\tau \pm (t-t'))]}{\pi(\tau \pm (t-t'))} \rightarrow \delta(t' - (t \pm \tau)), \tag{32}$$

$$\frac{1-\cos[\omega_M(\tau \pm (t-t'))]}{\pi\omega_M(\tau \pm (t-t'))^2} \rightarrow \delta(t' - (t \pm \tau)), \tag{33}$$

and

$$\frac{d^2}{dt'^2} \frac{\sin[\omega_M(\tau \pm (t-t'))]}{\pi(\tau \pm (t-t'))} \rightarrow \frac{d^2}{dt'^2} \delta(t' - (t \pm \tau)). \tag{34}$$

In addition, we take into account that the integration of the product of an odd function and an even function in the sense of principal value gives zero in our case. As one can see, the sign-changing functions are odd relative to the point  $t' = t - \tau$  in the above expressions for integrals (27), (28), and (31) with  $t > \tau$ , where  $\tau = \frac{r_{ij}}{c}$  is the time for light to pass the distance between the two atoms  $i$  and  $j$ . Furthermore, if a singular point is outside the region of integration, the integral value can be disregarded due to the “very fast” oscillating integrand and the above-limited norm for the time-dependent atomic operators  $|\alpha\rangle\langle\alpha|_i(t)$  and  $|\beta\rangle\langle\beta|_j(t')$  with a quite slowly changing functions  $f(t') = \tilde{f}(\hat{\rho}_{\alpha\alpha}^i(t'), r_{ij}(t'))$  during the time interval  $t$ . We suppose that the atomic positions change negligibly for the time interval equal to the minimum period  $\frac{2\pi}{\omega_{\max}}$ . Such situations arise, for example, for the integrals like the following ones:

$$\lim_{\omega_M \rightarrow \infty} \mathcal{P} \int_0^t dt' \frac{1-\cos[\omega_M(\tau \pm (t-t'))]}{\pi\omega_M(\tau \pm (t-t'))^3} \Phi(t') \rightarrow 0, \tag{35}$$

$$\lim_{\omega_M \rightarrow \infty} \mathcal{P} \int_0^t dt' \frac{\cos[\omega_M(\tau \pm (t-t'))]}{\pi\omega_M(\tau \pm (t-t'))} \Phi(t') \rightarrow 0, \tag{36}$$

and

$$\lim_{\omega_M \rightarrow \infty} \mathcal{P} \int_0^t dt' \frac{\sin[\omega_M(\tau \pm (t-t'))]}{(\tau \pm (t-t'))^2} \Phi(t') \rightarrow 0, \tag{37}$$

where  $\mathcal{P}$  stands for the principal value of the integral, and the symbol  $\Phi(t')$  is instead of  $f(t') |\alpha\rangle\langle\alpha|_i(t) |\beta\rangle\langle\beta|_j(t')$ .

As a result, we can use the following formulas:

$$I_0\left(\frac{r_{ij}}{c}, t-t'\right) \rightarrow -\frac{i\pi}{2} \left\{ \delta\left(t'-\left(t-\frac{r_{ij}}{c}\right)\right) - \delta\left(t'-\left(t+\frac{r_{ij}}{c}\right)\right) \right\}, \quad (38)$$

$$I_1\left(\frac{r_{ij}}{c}, t-t'\right) \rightarrow 0, \quad (39)$$

and, finally,

$$I_2\left(\frac{r_{ij}}{c}, t-t'\right) \rightarrow \frac{i\pi}{2} \left\{ \frac{\partial^2}{\partial t'^2} \delta\left(t'-\left(t-\frac{r_{ij}}{c}\right)\right) - \frac{\partial^2}{\partial t'^2} \delta\left(t'-\left(t+\frac{r_{ij}}{c}\right)\right) \right\}. \quad (40)$$

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#### ОТРИМАННЯ МОДЕЛЬНОГО ГАМІЛЬТОНІАНА ГРАНИЦІ “КОРОТКИХ ЧАСОВИХ ІНТЕРВАЛІВ”

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#### Резюме

Досліджено гамільтоніан, який описує нерелятивістську систему  $N$  частинок (атомів чи молекул), що взаємодіють із квантованим електромагнітним полем. Показано, що канонічне перетворення такого гамільтоніана, яке залишає у початковий момент часу лише оператори поля, для достатньо коротких проміжків часу не генерує поля, що обернено пропорційне першому і другому ступеню відстаней між частинками, а також розкриває певні колективні ефекти, включаючи захоплення електромагнітного поля системою частинок.