DYNAMICAL AND MACROSCOPIC DESCRIPTIONS OF THE STRONGLY DRIVEN GAS MEDIUM OF INTERACTING ATOMS

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This paper is devoted to constructing the microscopic and macroscopic theories of a system of N interacting two-level atoms coupled with a strong near resonant pumping field and a weak probe electromagnetic one. Microscopic kinetic equations for the density matrix elements of N-atom states including atomic motion are deduced with regard for the atom-field and atom-atom interactions. The corresponding macroscopic kinetics is built for the one- and two-particle density matrix distribution functions. The self-consistent system of macroscopic one-particle equations for the averaged density matrix elements along with the Maxwell equations allow us to describe the emission and absorption properties of the system and to explain the dependence of the optical properties on the particle density in terms of the "long-range" dipole-dipole interaction between the atoms.

1. Introduction

Here, we will construct the microscopic and macroscopic theories of a system of N interacting two-level atoms coupled with a strong near resonant pumping field and weak probe electromagnetic one. A theoretical investigation of the medium of "long-range" interacting particles placed in a driving strong coherent electromagnetic field needs to use complex mathematical tools and to seek a suitable model for the system. To develop a macroscopic model, we construct the exact microscopic evolution equations for an N-particle system interacting with near resonant fields.

The importance of many-particle effects for the optical properties of quite dense atomic gases were experimentally revealed. For instance, in [1, 2], the dipole-dipole spectral broadening of a resonance line was investigated in a dense rubidium vapor in the presence of a far-detuned pump beam. The experimental evidences of the influence of the atom-atom interaction on spectral characteristics can be found in [3] and, for example, [4–6]). The theoretical description of the effects including the spectral narrowing, spectral broadening, and radiation trapping caused by the long-range and short-range ("hard core" collisions) atom-atom interactions starts its development approximately from the second half of the twentieth century (see, e.g., [7–14]), by reaching the modern state, for example, in [15–17]. In the current investigation, we emphasize on the influence of the long-range interactions on line shapes, which was not previously explained theoretically.

Many techniques for deriving the master equation are in use today, but most of them are limited to the case of the non-interacting atoms (see, e.g., [18, 19–26]) or frozen positions for each atom (see, e.g., [27–31]). A review of the relatively early developments in the theory of binary collisions forming specific spectral profiles is presented in [32] with respect to the simplified master equations for one-particle density matrix elements, including some phenomenological items with regard for the atomic transitions at short-range collisions.

Other techniques, based on the Green's function method for non-equilibrium systems, under certain restrictions allow one to derive the kinetic equation for the density matrix of two-level particles [17]. The impact theory can be developed from the evolution equations for the quantum Green's functions under special simplifying assumptions about the correlations in the binary approximation (see [14]). Based on the appropriate formulation of the correlation functions (as in [7, 9, 10]), the impact approximation is an alternative to the density matrix formalism. The careful introduction of the binary collision integral, describing the intrinsic transitions in atoms during collisions, for the use in kinetic equations is the central moment in the impact theory (see [13, 14]). In our macroscopic theory, the items responsible for atomic transitions during short-range interactions are simplified in the relaxation approximation. This means that we represent the corresponding "short-range" part of the collision integral through the change of the density matrix relative to its equilibrium value related to the appropriate relaxation time (the details are explained and argued throughout the text). As in the impact theory, we neglect the space correlation between any two collided particles after a time interval much bigger than the short-range collision time. The constructed system of microscopic evolution equations allows us to obtain any approximation in the sense of many-body correlations, when the long-range interaction is taken into account. For example, we provide the derivation of the macroscopic evolution equations involving two-particle density matrix elements that are not available in the literature. Here, we restrict the description to the case of the absence of two-particle space correlations, but our microscopic theory allows us to develop the macroscopic description with regard for possible correlations. In comparison with the present paper, a number of works like [11, 12] developed the theory for equilibrium statistical systems within the Green's function method, by neglecting any kinetic processes in the medium of interacting particles.

Using a model Hamiltonian with the quantized optical pumping and probing electromagnetic fields and the quantized atomic motion, the idea of a collective atomic recoil laser (CARL) including the atomic dipole-dipole interaction was proposed in [33]. But, probably because of the already mentioned mathematical difficulties, the analysis of the possibility of CARL in a dense atomic vapor used a too simplified semiclassical system of equations for each atom. The huge amount of the semiclassical equations corresponding to the long-range many-particle dipole-dipole interactions cannot be used for an analysis. No macroscopic theory was developed in [33]. To make a system description, the authors proposed to carry out an appropriate simulation under certain conditions. It was revealed that the long-range interatomic interaction can play the "negative role" in the lasing process. This was referred to the non-coherent characteristic of collective atomic recoil motions. In our opinion, the results can be used for quite "cold" gases. In addition, as we showed in another paper, an interatomic interaction can act in different manner on the optical properties. For example, the presence of the dipole-dipole interaction can reduce or increase the "lasing ability" in quite "hot" atoms and become the cause for the ordered population and polarization distributions. We also propose a complete microscopic-macroscopic theory of the system, making a possibility to derive an analytic description.

In [34], the nonlinear dynamics of an open quantum system is treated for an arbitrary number of two-level non-colliding atoms coupled with a classical polychromatic field and one quantized mode of this field, but without consideration of the dipole-dipole interaction. A deeper analysis of the same system, adding non-adiabatic transitions, was provided in [35]. In [36], a similar analytical method, based on the generalized Jaynes–Cummings model, was used. That work discussed some characteristics of light in a two-mode cavity with the possibility of resonant two-photon transitions for non-interacting fermions, but did not include dissipative processes.

In this paper, we study the N-atom ensemble with regard for the possibility of a two-photon excitation or decay (jumps) involving a pair of atoms interacting with the dipole-dipole coupling in the strong-field regime. For comparison, the previous theoretical work [37] described the evolution of two dipole-dipole interacting atoms in a vacuum with only one atom being initially excited; work [31] 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 [29, 30] followed the approximations of [27, 28], just adding an additional state to the model corresponding to two simultaneously excited atoms. They examined possible two-photon jumps, which is different from our description of "jumps" (we model the dipole-dipole interaction by an operator keeping the appropriate pairs of atomic transition operators that are neglected in the approach in [27]: see the more detailed explanation in the next section).

As compared with works like [27, 37], where the well-localized atoms were investigated, we consider the interaction of atoms with a quantum radiation bath in the model describing the dipole-dipole coupling between atoms. The model is based on certain radiative effects, including the radiation trapping. We formulate the evolution equations for the system in terms of averaged one- and two-particle density matrix elements. The obtained macroscopic description allows us to examine the system of atoms acting as a whole and to compute the emission and absorption properties of the medium in a straightforward manner in view of certain statistical properties.

2. Main Statements

We start from the Schrödinger equation for the N-atom state

$$i\hbar\frac{\partial}{\partial t}|\Psi\rangle = \hat{\mathcal{H}}|\Psi\rangle \quad \text{with} \quad \hat{\mathcal{H}} = \hat{\mathrm{H}} + \hat{\Gamma}, \tag{1}$$

where $\hat{\mathrm{H}}$ is a part of the Hamiltonian that involves both the interaction of atoms with the electromagnetic field and the dipole-dipole interaction between atoms:

$$\hat{\mathrm{H}} = \hbar\omega_a \sum_{i=1}^{N} \sigma_i^+\sigma_i + \hbar\omega_b \sum_{i=1}^{N} \sigma_i\sigma_i^+ -$$

$$-\sum_{i=1}^{N}\left(\wp_{ab}^i\sigma_i^+ + \wp_{ba}^i\sigma_i\right)\cdot\mathcal{E}(t,\mathbf{r}_i) + \frac{1}{2}\frac{1}{4\pi\epsilon_0}\sum_{i,j;i\neq j}^{N,N}\frac{1}{|\mathbf{r}_{ij}|^3}\times$$

$$\times\Bigg[\left(\wp_{ab}^i\sigma_i^+ + \wp_{ba}^i\sigma_i\right)\cdot\left(\wp_{ab}^j\sigma_j^+ + \wp_{ba}^j\sigma_j\right) -$$

$$-3\hat{r}_{ij}\cdot\left(\wp_{ab}^i\sigma_i^+ + \wp_{ba}^i\sigma_i\right)\hat{r}_{ij}\cdot\left(\wp_{ab}^j\sigma_j^+ + \wp_{ba}^j\sigma_j\right)\Bigg]. \tag{2}$$

Here, $\sigma_i^+ = |a\rangle\langle b|_i$ and $\sigma_i = |b\rangle\langle a|_i$ are the excited state creation and annihilation operators for the i-th atom, $i = 1,\ldots,N$, respectively; N is the number of atoms; a and b denote the excited and ground states of an atom; $\mathbf{r}_i$ is the position of the i-th atom; $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$, $\hat{r}_{ij} = (\mathbf{r}_i - \mathbf{r}_j)/|\mathbf{r}_i - \mathbf{r}_j|$ is the unit vector pointing from atom j to atom i. The off-diagonal dipole matrix elements are defined by terms $\left(\wp_{ab}^i\right)^* = \wp_{ba}^i = \langle b|\hat{\mu}|a\rangle_i$, $i = 1,\ldots,N$; where $\hat{\mu}$ is the dipole operator for the atom.

The external electromagnetic field $\mathcal{E}(t,\mathbf{r}_i)$ at the point $\mathbf{r}_i$ includes a pump field, a probe field with close frequencies, and the far-field radiation from atoms. This can be presented in the form

$$\mathcal{E}(t,\mathbf{r}_i) = \mathbf{E}(t,\mathbf{r}_i) + \mathbf{E}^*(t,\mathbf{r}_i). \tag{3}$$

In the case where only the "pump" field is taken into account, we have $\mathbf{E}(t,\mathbf{r}) = \mathbf{E}_0 e^{-i(\omega t - \mathbf{k}\cdot\mathbf{r})}$. Here, ω is the pump frequency, and $\mathbf{k}$ is its wave vector.

In our approximation, the state vector $|\Psi\rangle$ has the form

$$|\Psi\rangle = \sum_{\alpha=(\alpha_1\ldots\alpha_N)}^{2^N} C_\alpha(t,\mathbf{X})|\Psi\rangle_\alpha$$

with

$$\{|\Psi\rangle_\alpha\} = \{|\alpha_1\ldots\alpha_i\ldots\alpha_N\rangle, \alpha_i = (a,b)\}, \tag{4}$$

where $\mathbf{X} = (\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N)$ are the actual atomic positions. The coefficient $C_\alpha(t,\mathbf{X})$ corresponds to the probability amplitude to find the system in a state α with defined N intrinsic atomic states (in the set α) and coordinates $\mathbf{X}\pm d\mathbf{X}$ at the moment of time $t \pm dt$. Therefore, generally speaking, the state vector also describes the correlations between all N atoms.

The part of the Hamiltonian, which describes the dipole-dipole interaction between all pairs of N atoms, is written with the following assumptions. First, all binary combinations of atomic operators σ_i and σ_j^+, like $\sigma_i^+\sigma_j^+$ and $\sigma_i\sigma_j$, with $i \neq j$, are included in the way shown above. This can be obtained on a "quite short time scale" by eliminating the time-dependent field operators in Hamiltonian (3) and by using the canonical transformation (7) in [27]. Next, the average distance between two atoms is assumed to be smaller than a sixth of the pump (and signal) wavelength. In addition, we consider the average distance between atoms to be many times bigger than the effective atomic size (diameter). Accordingly, the summation over pairs of atoms appearing in the Schrödinger equation has to take into account only such pairs of particles having separations less than the wavelength of a pump field over 2π (inverse of the wave vector). In this approximation, the dipole radiation from more distant atoms is included in the definition of the external electromagnetic field $\mathcal{E}(t,\mathbf{r}_i)$.

$\hat{\Gamma}$ is a part of the Hamiltonian $\hat{\mathcal{H}}$, which describes the damping effects. In our approximation, we neglect the collective atomic recoil, even though it can be present in the experiment. It is supposed that the time scale for the buildup of the gain is of the order of the spontaneous emission time (e.g., < 20 ns, as in [3]), so that collision processes and multiple spontaneous recoils (though possibly not stimulated recoils) play a negligible role. An important role in defining the system behavior belongs to a long-range interaction, which is treated separately from the introduced "damping" operator $\hat{\Gamma}$ in the model Hamiltonian. The model operator $\hat{\Gamma}$ is responsible for the damping of atomic states during atomic "hard core" collisions and under the interaction with the radiation bath (including vacuum fluctuations).

On a theoretical basis, the quantum recoil effects can be eliminated, as described in the recent work [37], because they require that the atomic motion on the time scale of spontaneous emission be small compared to the time taken to move a wavelength distance, as, otherwise, the Rabi frequency cannot be considered a constant (see works [29, 30, 37]). The contribution to the relaxation rates of collective states due to atomic "hard core" collisions and the interaction with the radiation bath is taken into account, by using the phenomenological decay coefficients in the kinetic equations given below. This model of decay guarantees the normalization of the system state vector and justifies the use of averaged one-particle states. In the section "Proof," we derive the properties of the damping operator $\hat{\Gamma}$ averaged over the atomic quantum states, which is the phenomenological damping.

Further, we do not quantize the motion of the atomic center of mass, due to the approximation of hot gas, but treat it semiclassically.

In the limit of a strong pump beam, we set $\wp^i_{ab} = \wp^i_{ba} = \wp\hat{\wp}$, $i = 1, \ldots, N$ (here, $\hat{\wp}$ is the unit vector parallel to the direction of the off-diagonal dipole matrix element), and assume a linear polarization of the pump, so that the off-diagonal dipole matrix elements follow the external pump polarization, by either reinforcing or cancelling its amplitude $\mathbf{E}_0$. Then the following microscopic evolution equations can be derived (see section "Proof"):

$$\frac{\partial}{\partial t}\rho^i_{aa}(t,\mathbf{X}) = \lambda_a - \gamma\rho^i_{aa}(t,\mathbf{X}) +$$

$$+\frac{i}{\hbar}\left(\rho^i_{ba}(t,\mathbf{X})\wp^i_{ab}\cdot\mathbf{E}(\mathbf{r}_i,t) - \rho^i_{ab}(t,\mathbf{X})\wp^i_{ba}\cdot\mathbf{E}^*(\mathbf{r}_i,t)\right) -$$

$$-\frac{1}{4\pi\epsilon_0}\frac{i}{\hbar}\sum_{j;j\neq i}^{N}\left\{\left[\rho^{i;j}_{ba;ba}(t,\mathbf{X}) + \rho^{i;j}_{ba;ab}(t,\mathbf{X}) -\right.\right.$$

$$\left.\left.-\rho^{i;j}_{ab;ab}(t,\mathbf{X}) - \rho^{i;j}_{ab;ba}(t,\mathbf{X})\right]Q^{ij}_{ab}(\mathbf{r}_{ij})\right\}; \tag{5}$$

$$\frac{\partial}{\partial t}\rho^i_{bb}(t,\mathbf{X}) = \lambda_b - \gamma\rho^i_{bb}(t,\mathbf{X}) -$$

$$-\frac{i}{\hbar}\left(\rho^i_{ba}(t,\mathbf{X})\wp^i_{ab}\cdot\mathbf{E}(\mathbf{r}_i,t) - \rho^i_{ab}(t,\mathbf{X})\wp^i_{ba}\cdot\mathbf{E}^*(\mathbf{r}_i,t)\right) +$$

$$+\frac{1}{4\pi\epsilon_0}\frac{i}{\hbar}\sum_{j;j\neq i}^{N}\left\{\left[\rho^{i;j}_{ba;ba}(t,\mathbf{X}) +\right.\right.$$

$$\left.\left.+\rho^{i;j}_{ba;ab}(t,\mathbf{X}) - \rho^{i;j}_{ab;ab}(t,\mathbf{X}) - \rho^{i;j}_{ab;ba}(t,\mathbf{X})\right]Q^{ij}_{ab}(\mathbf{r}_{ij})\right\}; \tag{6}$$

$$\frac{\partial}{\partial t}\rho^i_{ba}(t,\mathbf{X}) = -\gamma_{ba}\rho^i_{ba}(t,\mathbf{X}) + i\omega_0\rho^i_{ba}(t,\mathbf{X}) +$$

$$+\frac{i}{\hbar}\left(\rho^i_{aa}(t,\mathbf{X})\wp^i_{ba} - \rho^i_{bb}(t,\mathbf{X})\wp^i_{ab}\right)\cdot\mathbf{E}^*(\mathbf{r}_i,t) -$$

$$-\frac{1}{4\pi\epsilon_0}\frac{i}{\hbar}\sum_{j;j\neq i}^{N}\left\{\left[\rho^{i;j}_{aa;ba}(t,\mathbf{X}) + \rho^{i;j}_{aa;ab}(t,\mathbf{X}) -\right.\right.$$

$$\left.\left.-\rho^{i;j}_{bb;ab}(t,\mathbf{X}) - \rho^{i;j}_{bb;ba}(t,\mathbf{X})\right]Q^{ij}_{ab}(\mathbf{r}_{ij})\right\}; \tag{7}$$

$$m_i\frac{d^2}{dt^2}\mathbf{r}_i(t) = -\frac{\partial}{\partial\mathbf{r}_i}\left\{-\left(\rho^i_{ba}(t,\mathbf{X})\wp^i_{ba}\cdot\mathbf{E}(\mathbf{r}_i,t) +\right.\right.$$

$$\left.+\rho^i_{ab}(t,\mathbf{X})\wp^i_{ab}\cdot\mathbf{E}^*(\mathbf{r}_i,t)\right) +$$

$$+\frac{1}{4\pi\epsilon_0}\sum_{j;j\neq i}^{N}\left\{\left[\rho^{i;j}_{ba;ba}(t,\mathbf{X}) + \rho^{i;j}_{ba;ab}(t,\mathbf{X}) +\right.\right.$$

$$\left.\left.\left.+\rho^{i;j}_{ab;ab}(t,\mathbf{X}) + \rho^{i;j}_{ab;ba}(t,\mathbf{X})\right]Q^{ij}_{ab}(\mathbf{r}_{ij})\right\}\right\}. \tag{8}$$

Here, we used the notation

$$Q^{ij}_{ab}(\mathbf{r}_{ij}) = \left[\wp^i_{ab}\wp^j_{ab} - 3(\wp^i_{ab}\hat{r}_{ij})(\wp^j_{ab}\hat{r}_{ij})\right]\frac{1}{|\mathbf{r}_{ij}|^3}. \tag{9}$$

The atomic resonant frequency is defined as $\omega_0 = \omega_a - \omega_b$ and is, generally saying, a function of the atomic velocity because of the Doppler effect, so that each atom has own resonant frequency (the notation is kept without an atomic index here); $\lambda_a = \gamma\overline{n}^{(0)}_a$, $\lambda_b = \gamma\overline{n}^{(0)}_b$; $\overline{n}^{(0)}_a + \overline{n}^{(0)}_b = 1$, where $\overline{n}^{(0)}_a$ and $\overline{n}^{(0)}_b$ are the equilibrium solution of the equations above without external electromagnetic fields and without dipole-dipole interaction between the atoms. In accordance with our model, we use the phenomenological decay coefficients γ and γ_{ba} to include the spontaneous decay, collisions, and other interactions with the external environment. Here and further in the following text for simplicity, we count no difference between $\wp^i_{ba}$ and $\wp^i_{ab}$, or, in other words, $\wp^i_{ba} = \left(\wp^i_{ba}\right)^*$. Moreover, we used the following definitions for the microscopic N-particle density matrix elements:

$$\rho^i_{aa} = \sum_{\beta}^{2^N} C^*_{\beta_1\ldots a_i\ldots\beta_N}C_\beta\,\delta_{\beta_i a};$$

$$\rho^{i}_{ab}=\sum_{\beta}^{2^N} C^{*}_{\beta_1\ldots b_i\ldots\beta_N} C_{\beta}\,\delta_{\beta_i a};$$

$$\rho^{i}_{bb}=\sum_{\beta}^{2^N} C^{*}_{\beta_1\ldots b_i\ldots\beta_N} C_{\beta}\,\delta_{\beta_i b};\qquad(10)$$

$$\rho^{i;j}_{ab;ba}=\sum_{\beta}^{2^N} C^{*}_{\beta_1\ldots b_i\ldots a_j\ldots\beta_N} C_{\beta}\,\delta_{\beta_i a}\,\delta_{\beta_j b};$$

$$\rho^{i;j}_{aa;ba}=\sum_{\beta}^{2^N} C^{*}_{\beta_1\ldots a_i\ldots a_j\ldots\beta_N} C_{\beta}\,\delta_{\beta_i a}\,\delta_{\beta_j b};$$

$$\rho^{i;j}_{ba;ba}=\sum_{\beta}^{2^N} C^{*}_{\beta_1\ldots a_i\ldots a_j\ldots\beta_N} C_{\beta}\,\delta_{\beta_i b}\,\delta_{\beta_j b}.\qquad(11)$$

Here, $\beta=(\beta_1\ldots\beta_i\ldots\beta_N)$, $\beta_i=(a,b)$ and $\delta_{\beta_j b}$ is the Kronecker delta. The coefficients $C_\beta=C_\beta(t,\mathbf{X})$ are defined in (4).

Averaging the above "N-particle" equations with an ensemble distribution and using the symmetry of the ensemble relative to the permutation of any two atoms, the chain of one-, two-, three-, ..., N-particle kinetic equations can be obtained. Then, assuming that a spatial correlation between any two atoms quickly vanishes with time (mathematically it is written as the expressions (53)), we can obtain the following system of one-particle self-consistent macroscopic evolution equations from the corresponding N-particle microscopic equations (5)–(8) or the one- and two-particle macroscopic equations (43)–(46) based on the model Hamiltonian (see the detailed definitions and derivations of the equations in the section "Proof") with the notation $\chi=\frac{1}{4\pi\varepsilon_0}(N-1)\wp^2$:

$$\frac{\partial}{\partial t}\rho_{aa}(t,\mathbf{r})=\lambda_a\frac{n(\mathbf{r})}{N}-\gamma\rho_{aa}(t,\mathbf{r})+$$
$$+\frac{i}{\hbar}\wp\left(\rho_{ba}(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}(t,\mathbf{r})-\rho_{ab}(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}^{*}(t,\mathbf{r})\right)+$$
$$+\frac{2}{\hbar}\chi\int d\mathbf{r}'\left\{2\,\mathrm{Im}\left(\rho_{ba}(t,\mathbf{r})\right)\mathrm{Re}\left(\rho_{ba}(t,\mathbf{r}')\right)Q\left(\mathbf{r},\mathbf{r}'\right)\right\},\qquad(12)$$

where $\rho_{aa}(t,\mathbf{r})$ is the excited state one-particle probability density given by the averaged density matrix element, which describes the probability to find an atom at the time $t\pm dt$ and the position $\mathbf{r}\pm d\mathbf{r}$ in the excited state a independently of whether the other atoms in the vapor cell (volume V) are in the excited a or ground b state. We have

$$\frac{\partial}{\partial t}\rho_{bb}(t,\mathbf{r})=\lambda_b\frac{n(\mathbf{r})}{N}-\gamma\rho_{bb}(t,\mathbf{r})-$$
$$-\frac{i}{\hbar}\wp\left(\rho_{ba}(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}(t,\mathbf{r})-\rho_{ab}(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}^{*}(t,\mathbf{r})\right)-$$
$$-\frac{2}{\hbar}\chi\int d\mathbf{r}'\left\{2\,\mathrm{Im}\left(\rho_{ba}(t,\mathbf{r})\right)\mathrm{Re}\left(\rho_{ba}(t,\mathbf{r}')\right)Q\left(\mathbf{r},\mathbf{r}'\right)\right\},\qquad(13)$$

where $\rho_{bb}(t,\mathbf{r})$ is the ground state one-particle probability density given by the averaged density matrix element, which describes the probability to find an atom at the time $t\pm dt$ and the position $\mathbf{r}\pm d\mathbf{r}$ in the ground state b independently of whether the other atoms in the vapor cell (volume V) are in the excited a or ground b state. We obtain

$$\frac{\partial}{\partial t}\rho_{ba}(t,\mathbf{r})=-\gamma_{ba}\rho_{ba}(t,\mathbf{r})+i\overline{\omega}_0\rho_{ba}(t,\mathbf{r})+$$
$$+\frac{i}{\hbar}\left(\rho_{aa}(t,\mathbf{r})-\rho_{bb}(t,\mathbf{r})\right)\wp\hat{\wp}\cdot\mathbf{E}^{*}(t,\mathbf{r})-$$
$$-\frac{2i}{\hbar}\chi\int d\mathbf{r}'\left\{\left(\rho_{aa}(t,\mathbf{r})-\rho_{bb}(t,\mathbf{r})\right)\times\right.$$
$$\left.\times\mathrm{Re}\left(\rho_{ba}(t,\mathbf{r}')\right)Q\left(\mathbf{r},\mathbf{r}'\right)\right\},\qquad(14)$$

where $\rho_{ba}(t,\mathbf{r})$ is the averaged "off-diagonal" density matrix element, which determines a polarization of the gas medium.

Finally, the equation for the quasiclassical particle translational motion describing a change of the averaged atomic velocity $\mathbf{u}(t,\mathbf{r})$ is as follows:

$$m\frac{\partial}{\partial t}\mathbf{u}(t,\mathbf{r})=-m\left(\mathbf{u}(t,\mathbf{r})\cdot\frac{\partial}{\partial\mathbf{r}}\right)\mathbf{u}(t,\mathbf{r})-$$
$$-\frac{\partial}{\partial\mathbf{r}}\left\{-\wp\left(\rho_{ba}(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}(t,\mathbf{r})+\rho_{ab}(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}^{*}(t,\mathbf{r})\right)+\right.$$
$$\left.+2\chi\int d\mathbf{r}'\left\{2\,\mathrm{Re}\left(\rho_{ba}(t,\mathbf{r})\right)\mathrm{Re}\left(\rho_{ba}(t,\mathbf{r}')\right)Q\left(\mathbf{r},\mathbf{r}'\right)\right\}\right\}.\qquad(15)$$

Here,

$$Q\left(\mathbf{r},\mathbf{r}'\right)=\frac{\hat{\wp}\cdot\hat{\wp}'}{\left|\mathbf{r}-\mathbf{r}'\right|^{3}}-3\frac{\left(\hat{\wp}\cdot(\mathbf{r}-\mathbf{r}')\right)\left(\hat{\wp}'\cdot(\mathbf{r}-\mathbf{r}')\right)}{\left|\mathbf{r}-\mathbf{r}'\right|^{5}};\qquad(16)$$

$\overline{\omega}_0$ is the atomic "resonant" frequency averaged over the ensemble. This averaged value can depend on time and coordinates, as shown in the subsection "From microscopic description to macroscopic" of the next section. The parameters $\lambda_{a,b}$ like the previous microscopic case describe the non-averaged density matrix elements for a system of non-interacting atoms without external electromagnetic field. These are defined in the next section. By $n(\mathbf{r})$, we denote the number of atoms per unit volume.

To take the long-range atom-atom interactions into account, we need to use Maxwell's equations for the macroscopic electromagnetic field propagating in the medium:

$$\nabla \times \nabla \times \mathbf{E}(t, \mathbf{r}) + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E}(t, \mathbf{r}) = -\mu_0 \frac{\partial^2}{\partial t^2} \mathbf{P}(t, \mathbf{r}), \quad (17)$$

where the polarization $\mathbf{P}(t, \mathbf{r})$ of the system per unit volume is given by

$$\mathbf{P}(t, \mathbf{r}) = N \wp \hat{\wp} \left(\rho_{ba}(t, \mathbf{r}) + \rho_{ab}(t, \mathbf{r}) \right), \quad (18)$$

and c is the light speed in the medium.

Here, we need to keep in mind the necessity to use one mutual spatial scale for the three terms: the macroscopic one-particle distribution, the macroscopic density matrix, and the macroscopic electromagnetic field. Accordingly, the the macroscopic density matrix elements $\rho_{aa}(t, \mathbf{r})$, $\rho_{ba}(t, \mathbf{r})$, and $\rho_{ba}(t, \mathbf{r})$ are not pure atom states but include the action of the pump field and the ensemble averaging.

3. Proof

Here, we derive the evolution equations for the state amplitudes of the N-atom system. The evolution equations for the density matrix and coordinates of an atom are first obtained. Then, after the microscopic description of the system, we explain the method of transforming these equations into a self-consistent system of one-particle macroscopic kinetic equations.

3.1. Evolution equations for the state amplitudes of the system

We multiply the left- and right-hand sides of Eq. (1) by $\langle \Psi |_\beta = \langle \beta_1 \ldots \beta_i \ldots \beta_j \ldots \beta_N |$ and obtain

$$\frac{\partial}{\partial t} C_\beta(t, \mathbf{X}) = -i\omega_a \sum_{i=1}^{N} \sum_{\alpha}^{2^N} C_\alpha(t, \mathbf{X}) \langle \Psi |_\beta \sigma_i^+ \sigma_i | \Psi \rangle_\alpha -$$

$$-i\omega_b \sum_{i=1}^{N} \sum_{\alpha}^{2^N} C_\alpha(t, \mathbf{X}) \langle \Psi |_\beta \sigma_i \sigma_i^+ | \Psi \rangle_\alpha +$$

$$+\frac{i}{\hbar} \sum_{i=1}^{N} \sum_{\alpha}^{2^N} C_\alpha(t, \mathbf{X}) \left(\wp_{ab}^i \langle \Psi |_\beta \sigma_i^+ | \Psi \rangle_\alpha + \right.$$

$$\left. + \wp_{ba}^i \langle \Psi |_\beta \sigma_i | \Psi \rangle_\alpha \right) \cdot \mathcal{E}(t, \mathbf{r}_i) -$$

$$-\frac{1}{2} \frac{1}{4\pi\epsilon_0} \frac{i}{\hbar} \sum_{i,j;i\neq j}^{N,N} \sum_{\alpha}^{2^N} C_\alpha(t, \mathbf{X}) \left[\wp_{ab}^i \cdot \wp_{ab}^j \langle \Psi |_\beta \sigma_i^+ \sigma_j^+ | \Psi \rangle_\alpha + \right.$$

$$+ \wp_{ab}^i \cdot \wp_{ba}^j \langle \Psi |_\beta \sigma_i^+ \sigma_j | \Psi \rangle_\alpha +$$

$$+ \wp_{ba}^i \cdot \wp_{ab}^j \langle \Psi |_\beta \sigma_i \sigma_j^+ | \Psi \rangle_\alpha + \wp_{ba}^i \cdot \wp_{ba}^j \langle \Psi |_\beta \sigma_i \sigma_j | \Psi \rangle_\alpha -$$

$$-3 \left((\hat{r}_{ij} \cdot \wp_{ab}^i)(\hat{r}_{ij} \cdot \wp_{ab}^j) \langle \Psi |_\beta \sigma_i^+ \sigma_j^+ | \Psi \rangle_\alpha + \right.$$

$$+ (\hat{r}_{ij} \cdot \wp_{ab}^i)(\hat{r}_{ij} \cdot \wp_{ba}^j) \langle \Psi |_\beta \sigma_i^+ \sigma_j | \Psi \rangle_\alpha +$$

$$+ (\hat{r}_{ij} \cdot \wp_{ba}^i)(\hat{r}_{ij} \cdot \wp_{ab}^j) \langle \Psi |_\beta \sigma_i \sigma_j^+ | \Psi \rangle_\alpha +$$

$$\left. \left. + (\hat{r}_{ij} \cdot \wp_{ba}^i)(\hat{r}_{ij} \cdot \wp_{ba}^j) \langle \Psi |_\beta \sigma_i \sigma_j | \Psi \rangle_\alpha \right) \right] \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|^3} -$$

$$-\frac{i}{\hbar} \sum_{\alpha}^{2^N} C_\alpha(t, \mathbf{X}) \langle \Psi |_\beta \hat{\Gamma} | \Psi \rangle_\alpha. \quad (19)$$

After the substitution of the expressions for the averaged operators (24), (25) (see in the next subsection), we have

$$\frac{\partial}{\partial t} C_\beta(t, \mathbf{X}) = -i\omega_a \sum_{i=1}^{N} C_{\beta_1 \ldots a_i \ldots \beta_N} \delta_{\beta_i a} -$$

$$-i\omega_b \sum_{i=1}^{N} C_{\beta_1 \ldots b_i \ldots \beta_N} \delta_{\beta_i b} + \frac{i}{\hbar} \sum_{i}^{N} \left(C_{\beta_1 \ldots b_i \ldots \beta_N} \delta_{\beta_i a} \wp_{ab}^i + \right.$$

$$\left. + C_{\beta_1 \ldots a_i \ldots \beta_N} \delta_{\beta_i b} \wp_{ba}^i \right) \cdot \mathcal{E}(t, \mathbf{r}_i) -$$

$$-\frac{1}{2} \frac{1}{4\pi\epsilon_0} \frac{i}{\hbar} \sum_{i,j;i\neq j}^{N;N} \left[C_{\beta_1 \ldots b_i \ldots b_j \ldots \beta_N} \delta_{\beta_i a} \delta_{\beta_j a} \wp_{ab}^i \cdot \wp_{ab}^j + \right.$$

$$+ C_{\beta_1 \ldots b_i \ldots a_j \ldots \beta_N} \delta_{\beta_i a} \delta_{\beta_j b} \wp_{ab}^i \cdot \wp_{ba}^j +$$

$$+ C_{\beta_1 \ldots a_i \ldots b_j \ldots \beta_N} \delta_{\beta_i b} \delta_{\beta_j a} \wp_{ba}^i \cdot \wp_{ab}^j +$$

$$+ C_{\beta_1 \ldots a_i \ldots a_j \ldots \beta_N} \delta_{\beta_i b} \delta_{\beta_j b} \wp_{ba}^i \cdot \wp_{ba}^j -$$

$$-3 \left(C_{\beta_1 \ldots b_i \ldots b_j \ldots \beta_N} \delta_{\beta_i a} \delta_{\beta_j a} (\wp_{ab}^i \cdot \hat{r}_{ij})(\wp_{ab}^j \cdot \hat{r}_{ij}) + \right.$$

$$+ C_{\beta_1 \ldots b_i \ldots a_j \ldots \beta_N} \delta_{\beta_i a} \delta_{\beta_j b} (\wp_{ab}^i \cdot \hat{r}_{ij})(\wp_{ba}^j \cdot \hat{r}_{ij}) +$$

$$+ C_{\beta_1 \ldots a_i \ldots b_j \ldots \beta_N} \delta_{\beta_i b} \delta_{\beta_j a} (\wp_{ba}^i \cdot \hat{r}_{ij})(\wp_{ab}^j \cdot \hat{r}_{ij}) +$$

$$\left. \left. + C_{\beta_1 \ldots a_i \ldots a_j \ldots \beta_N} \delta_{\beta_i b} \delta_{\beta_j b} (\wp_{ba}^i \cdot \hat{r}_{ij})(\wp_{ba}^j \cdot \hat{r}_{ij}) \right) \right] \times$$

$$\times \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|^3} - \frac{i}{\hbar} \sum_{\alpha}^{2^N} C_\alpha(t, \mathbf{X}) \langle \Psi |_\beta \hat{\Gamma} | \Psi \rangle_\alpha. \quad (20)$$

3.2. Equation of motion for an atom

In the semiclassical approximation, the equation of motion for an atom has the following form **between collisions**:

$$m_i\frac{d^2}{dt^2}\mathbf{r}_i(t)=-\frac{\partial}{\partial\mathbf{r}_i}\langle\hat{\mathrm{H}}_i\rangle, \tag{21}$$

where

$$\hat{\mathrm{H}}_i=-\left(\wp^i_{ab}\sigma^+_i+\wp^i_{ba}\sigma_i\right)\cdot\mathcal{E}\left(t,\mathbf{r}_i\right)+\frac{1}{4\pi\epsilon_0}\times$$

$$\times\sum_{j;i\neq j}^{N}\frac{1}{|\mathbf{r}_i-\mathbf{r}_j|^3}\Big[\left(\wp^i_{ab}\sigma^+_i+\wp^i_{ba}\sigma_i\right)\cdot\left(\wp^j_{ab}\sigma^+_j+\wp^j_{ba}\sigma_j\right)-$$

$$-3\hat{r}_{ij}\cdot\left(\wp^i_{ab}\sigma^+_i+\wp^i_{ba}\sigma_i\right)\hat{r}_{ij}\cdot\left(\wp^j_{ab}\sigma^+_j+\wp^j_{ba}\sigma_j\right)\Big]. \tag{22}$$

Then we have

$$m_i\frac{d^2}{dt^2}\mathbf{r}_i(t)=-\frac{\partial}{\partial\mathbf{r}_i}\sum_{\beta}^{2^N}\sum_{\alpha}^{2^N}C^*_\beta(t,\mathbf{X})C_\alpha(t,\mathbf{X})\times$$

$$\times\Big\{\frac{1}{|\mathbf{r}_i-\mathbf{r}_j|^3}-\left(\wp^i_{ab}\langle\Psi|_\beta\sigma^+_i|\Psi\rangle_\alpha+\wp^i_{ba}\langle\Psi|_\beta\sigma_i|\Psi\rangle_\alpha\right)\times$$

$$\times\mathcal{E}\left(t,\mathbf{r}_i\right)+\frac{1}{4\pi\epsilon_0}\sum_{j;j\neq i}^{N}\Big[\wp^i_{ab}\cdot\wp^j_{ab}\langle\Psi|_\beta\sigma^+_i\sigma^+_j|\Psi\rangle_\alpha+$$

$$+\wp^i_{ab}\cdot\wp^j_{ba}\langle\Psi|_\beta\sigma^+_i\sigma_j|\Psi\rangle_\alpha+\wp^i_{ba}\cdot\wp^j_{ab}\langle\Psi|_\beta\sigma_i\sigma^+_j|\Psi\rangle_\alpha+$$

$$+\wp^i_{ba}\cdot\wp^j_{ba}\langle\Psi|_\beta\sigma_i\sigma_j|\Psi\rangle_\alpha-3\Big((\hat{r}_{ij}\cdot\wp^i_{ab})(\hat{r}_{ij}\cdot\wp^j_{ab})\times$$

$$\times\langle\Psi|_\beta\sigma^+_i\sigma^+_j|\Psi\rangle_\alpha+(\hat{r}_{ij}\cdot\wp^i_{ab})(\hat{r}_{ij}\cdot\wp^j_{ba})\times$$

$$\times\langle\Psi|_\beta\sigma^+_i\sigma_j|\Psi\rangle_\alpha+(\hat{r}_{ij}\cdot\wp^i_{ba})(\hat{r}_{ij}\cdot\wp^j_{ab})\langle\Psi|_\beta\sigma_i\sigma^+_j|\Psi\rangle_\alpha+$$

$$+(\hat{r}_{ij}\cdot\wp^i_{ba})(\hat{r}_{ij}\cdot\wp^j_{ba})\langle\Psi|_\beta\sigma_i\sigma_j|\Psi\rangle_\alpha\Big)\Big]\frac{1}{|\mathbf{r}_i-\mathbf{r}_j|^3}\Big\}. \tag{23}$$

After the substitution of the expressions for the averaged operators, we obtain

$$\sigma^+_i\sigma_i|\Psi\rangle_\alpha=\left(|a\rangle\langle a|_i\right)|\alpha_1\ldots\alpha_i\ldots\alpha_N\rangle=\delta_{a\alpha_i}|\Psi\rangle_\alpha;$$

$$\sigma^+_i|\Psi\rangle_\alpha=\left(|a\rangle\langle b|_i\right)|\alpha_1\ldots\alpha_i\ldots\alpha_N\rangle=\delta_{b\alpha_i}|\alpha_1\ldots a_i\ldots\alpha_N\rangle;$$

$$\langle\Psi|_\beta\sigma^+_i|\Psi\rangle_\alpha=\delta_{b\alpha_i}\delta_{\beta_1\alpha_1}\ldots\delta_{\beta_ia}\ldots\delta_{\beta_N\alpha_N};$$

$$\sigma_i|\Psi\rangle_\alpha=\left(|b\rangle\langle a|_i\right)|\alpha_1\ldots\alpha_i\ldots\alpha_N\rangle=\delta_{a\alpha_i}|\alpha_1\ldots b_i\ldots\alpha_N\rangle;$$

$$\langle\Psi|_\beta\sigma_i|\Psi\rangle_\alpha=\delta_{a\alpha_i}\delta_{\beta_1\alpha_1}\ldots\delta_{\beta_ib}\ldots\delta_{\beta_N\alpha_N}; \tag{24}$$

$$\sigma^+_i\sigma^+_j|\Psi\rangle_\alpha=\sigma^+_i\delta_{b\alpha_j}|\alpha_1\ldots a_j\ldots\alpha_N\rangle=$$

$$=\delta_{b\alpha_i}\delta_{b\alpha_j}|\alpha_1\ldots a_i\ldots a_j\ldots\alpha_N\rangle;$$

$$\langle\Psi|_\beta\sigma^+_i\sigma^+_j|\Psi\rangle_\alpha=$$

$$=\delta_{b\alpha_i}\delta_{b\alpha_j}\delta_{\beta_1\alpha_1}\delta_{\beta_2\alpha_2}\ldots\delta_{\beta_ia}\ldots\delta_{\beta_ja}\ldots\delta_{\beta_N\alpha_N};$$

$$\sigma^+_i\sigma_j|\Psi\rangle_\alpha=\sigma^+_i\delta_{a\alpha_j}|\alpha_1\ldots b_j\ldots\alpha_N\rangle=$$

$$=\delta_{b\alpha_i}\delta_{a\alpha_j}|\alpha_1\ldots a_i\ldots b_j\ldots\alpha_N\rangle;$$

$$\langle\Psi|_\beta\sigma^+_i\sigma_j|\Psi\rangle_\alpha=\delta_{b\alpha_i}\delta_{a\alpha_j}\delta_{\beta_1\alpha_1}\ldots\delta_{\beta_ia}\ldots\delta_{\beta_jb}\ldots\delta_{\beta_N\alpha_N};$$

$$\langle\Psi|_\beta\sigma_i\sigma^+_j|\Psi\rangle_\alpha=\delta_{a\alpha_i}\delta_{b\alpha_j}\delta_{\beta_1\alpha_1}\ldots\delta_{\beta_ib}\ldots\delta_{\beta_ja}\ldots\delta_{\beta_N\alpha_N};$$

$$\langle\Psi|_\beta\sigma_i\sigma_j|\Psi\rangle_\alpha=\delta_{a\alpha_i}\delta_{a\alpha_j}\delta_{\beta_1\alpha_1}\ldots\delta_{\beta_ib}\ldots\delta_{\beta_jb}\ldots\delta_{\beta_N\alpha_N};$$

$$\langle\Psi|_\beta\sigma^+_i\sigma_i|\Psi\rangle_\alpha=\delta_{a\alpha_i}\delta_{\beta_1\alpha_1}\ldots\delta_{\beta_ia}\ldots\delta_{\beta_N\alpha_N} \tag{25}$$

and the following equation of motion for atom i:

$$m_i\frac{d^2}{dt^2}\mathbf{r}_i(t)=-\frac{\partial}{\partial\mathbf{r}_i}\sum_{\beta}^{2^N}C^*_\beta(t,\mathbf{X})\times$$

$$\times\Big\{\frac{1}{|\mathbf{r}_i-\mathbf{r}_j|^3}-\left(C_{\beta_1\ldots b_i\ldots\beta_N}\delta_{\beta_ia}\wp^i_{ab}+C_{\beta_1\ldots a_i\ldots\beta_N}\delta_{\beta_ib}\wp^i_{ba}\right)\times$$

$$\times\mathcal{E}\left(t,\mathbf{r}_i\right)+\frac{1}{4\pi\epsilon_0}\sum_{j;j\neq i}^{N}\Big[C_{\beta_1\ldots b_i\ldots b_j\ldots\beta_N}\delta_{\beta_ia}\delta_{\beta_ja}\wp^i_{ab}\cdot\wp^j_{ab}+$$

$$+C_{\beta_1\ldots b_i\ldots a_j\ldots\beta_N}\delta_{\beta_ia}\delta_{\beta_jb}\wp^i_{ab}\cdot\wp^j_{ba}+$$

$$+C_{\beta_1\ldots a_i\ldots b_j\ldots\beta_N}\delta_{\beta_ib}\delta_{\beta_ja}\wp^i_{ba}\cdot\wp^j_{ab}+$$

$$+C_{\beta_1\ldots a_i\ldots a_j\ldots\beta_N}\delta_{\beta_ib}\delta_{\beta_jb}\wp^i_{ba}\cdot\wp^j_{ba}-$$

$$-3\Big(C_{\beta_1\ldots b_i\ldots b_j\ldots\beta_N}\delta_{\beta_ia}\delta_{\beta_ja}(\wp^i_{ab}\cdot\hat{r}_{ij})(\wp^j_{ab}\cdot\hat{r}_{ij})+$$

$$+C_{\beta_1\ldots b_i\ldots a_j\ldots\beta_N}\delta_{\beta_ia}\delta_{\beta_jb}(\wp^i_{ab}\cdot\hat{r}_{ij})(\wp^j_{ba}\cdot\hat{r}_{ij})+$$

$$+C_{\beta_1\ldots a_i\ldots b_j\ldots\beta_N}\delta_{\beta_ib}\delta_{\beta_ja}(\wp^i_{ba}\cdot\hat{r}_{ij})(\wp^j_{ab}\cdot\hat{r}_{ij})+$$

$$+C_{\beta_1\ldots a_i\ldots a_j\ldots\beta_N}\delta_{\beta_ib}\delta_{\beta_jb}(\wp^i_{ba}\cdot\hat{r}_{ij})(\wp^j_{ba}\cdot\hat{r}_{ij})\Big)\Big]\times$$

$$\times\frac{1}{|\mathbf{r}_i-\mathbf{r}_j|^3}\Big\}. \tag{26}$$

3.3. Evolution equations for the density matrix and coordinates of an atom

Now, we can write down expressions for the time derivative of the density matrix

$$\frac{\partial}{\partial t}\rho^i_{aa}(t,\mathbf{X}),\quad \frac{\partial}{\partial t}\rho^i_{ba}(t,\mathbf{X}),\quad \frac{\partial}{\partial t}\rho^i_{bb}(t,\mathbf{X}), \tag{27}$$

where

$$\rho^i_{aa}=\sum_{\beta}^{2^N}C^*_{\beta_1\ldots a_i\ldots\beta_N}C_\beta\,\delta_{\beta_i a},$$

$$\rho^i_{ab}=\sum_{\beta}^{2^N}C^*_{\beta_1\ldots b_i\ldots\beta_N}C_\beta\,\delta_{\beta_i a},$$

$$\rho^i_{bb}=\sum_{\beta}^{2^N}C^*_{\beta_1\ldots b_i\ldots\beta_N}C_\beta\,\delta_{\beta_i b}, \tag{28}$$

and make necessary transformations in the equations for the atomic coordinates. Note, here and in definitions (10) and (11), the above notations involving the Kronecker-delta mean the following:

$$\sum_{\beta}^{2^N}C^*_{\beta_1\ldots a_i\ldots\beta_N}C_\beta\,\delta_{\beta_i a}=\sum_{\beta}^{2^N}C^*_{\beta_1\ldots a_i\ldots\beta_N}C_{\beta_1\ldots a_i\ldots\beta_N};$$

$$\sum_{\beta}^{2^N}C^*_{\beta_1\ldots b_i\ldots\beta_N}C_\beta\delta_{\beta_i a}=\sum_{\beta}^{2^N}C^*_{\beta_1\ldots b_i\ldots\beta_N}C_{\beta_1\ldots a_i\ldots\beta_N};$$

$$\sum_{\beta}^{2^N}C^*_{\beta_1\ldots b_i\ldots\beta_N}C_\beta\,\delta_{\beta_i b}=\sum_{\beta}^{2^N}C^*_{\beta_1\ldots b_i\ldots\beta_N}C_{\beta_1\ldots b_i\ldots\beta_N}. \tag{29}$$

3.3.1. Normalization condition and the operator $\hat{\Gamma}$

To satisfy the normalization condition

$$\rho^i_{aa}(t,\mathbf{X})+\rho^i_{bb}(t,\mathbf{X})=1, \tag{30}$$

we accept such model "relaxation" operator $\hat{\Gamma}$ that

$$-\frac{i}{\hbar}\sum_{\beta}^{2^N}C^*_{\beta_1\ldots a_i\ldots\beta_N}\sum_{\alpha}^{2^N}C_\alpha\langle\Psi|_{\beta_1\ldots a_i\ldots\beta_N}\hat{\Gamma}|\Psi\rangle_\alpha+$$

$$+\frac{i}{\hbar}\sum_{\beta}^{2^N}C_{\beta_1\ldots a_i\ldots\beta_N}\sum_{\alpha}^{2^N}C^*_\alpha\left(\langle\Psi|_{\beta_1\ldots a_i\ldots\beta_N}\hat{\Gamma}|\Psi\rangle_\alpha\right)^*=$$

$$=\gamma_{bb}\rho^i_{bb}\left(t,\mathbf{X}\right)-\gamma_{aa}\rho^i_{aa}\left(t,\mathbf{X}\right). \tag{31}$$

By analogy, we have

$$-\frac{i}{\hbar}\sum_{\beta}^{2^N}C^*_{\beta_1\ldots b_i\ldots\beta_N}\sum_{\alpha}^{2^N}C_\alpha\langle\Psi|_{\beta_1\ldots b_i\ldots\beta_N}\hat{\Gamma}|\Psi\rangle_\alpha+$$

$$+\frac{i}{\hbar}\sum_{\beta}^{2^N}C_{\beta_1\ldots b_i\ldots\beta_N}\sum_{\alpha}^{2^N}C^*_\alpha\left(\langle\Psi|_{\beta_1\ldots b_i\ldots\beta_N}\hat{\Gamma}|\Psi\rangle_\alpha\right)^*=$$

$$=\gamma_{aa}\rho^i_{aa}\left(t,\mathbf{X}\right)-\gamma_{bb}\rho^i_{bb}\left(t,\mathbf{X}\right). \tag{32}$$

But,

$$-\frac{i}{\hbar}\sum_{\beta}^{2^N}C^*_{\beta_1\ldots a_i\ldots\beta_N}\sum_{\alpha}^{2^N}C_\alpha\langle\Psi|_{\beta_1\ldots b_i\ldots\beta_N}\hat{\Gamma}|\Psi\rangle_\alpha+$$

$$+\frac{i}{\hbar}\sum_{\beta}^{2^N}C_{\beta_1\ldots b_i\ldots\beta_N}\sum_{\alpha}^{2^N}C^*_\alpha\left(\langle\Psi|_{\beta_1\ldots a_i\ldots\beta_N}\hat{\Gamma}|\Psi\rangle_\alpha\right)^*=$$

$$=-\gamma_{ba}\rho^i_{ba}\left(t,\mathbf{X}\right). \tag{33}$$

3.3.2. Equations of motion for density matrix elements

To obtain the promised equation of motion, we substitut expression (20) into the formulae for partial derivatives introduced previously. For example,

$$\frac{\partial}{\partial t}\rho^i_{aa}(t,\mathbf{X})=\sum_{\beta}^{2^N}C_\beta\,\delta_{\beta_i a}\frac{\partial}{\partial t}C^*_{\beta_1\ldots a_i\ldots\beta_N}+$$

$$+\sum_{\beta}^{2^N}C^*_{\beta_1\ldots a_i\ldots\beta_N}\frac{\partial}{\partial t}C_\beta\,\delta_{\beta_i a}. \tag{34}$$

Consequently, using the results from previous subsections and the identity above, the system of dynamic (microscopic) equations (5)–(8) for atom i can be obtained in the rotating-wave approximation for the atom-field interaction.

3.4. From the microscopic description to the macroscopic one

Let $D\left(t,\mathbf{r}_1(t),\mathbf{r}_2(t),\ldots,\mathbf{r}_N(t)\right)=D\left(t,\mathbf{X}\right)$ be the probability density to find the system in a state with $\mathbf{X}\pm d\mathbf{X}$. Then the averaged probability density to find atom i in the excited state at the position $\mathbf{r}_i\pm d\mathbf{r}_i$ can be defined as

$$\overline{\rho^i_{aa}}\left(t,\mathbf{r}_i\right)=\int D(t,\mathbf{X})\rho^i_{aa}(t,\mathbf{X})\times$$

$$\times d\mathbf{r}_1 d\mathbf{r}_2 \, \ldots \, d\mathbf{r}_{i-1} d\mathbf{r}_{i+1} \, \ldots \, d\mathbf{r}_N. \tag{35}$$

To describe the system of atoms on different space- and time-scales, we develop a *kinetic approximation*, by introducing the splitting of the two-particle density matrix into a product of two one-particle density matrices,

$$\overline{\rho^{i;j}_{ab;ba}}(t, \mathbf{r}_i, \mathbf{r}_j) = \int D(t, \mathbf{X}) \, \rho^{i;j}_{ab;ba}(t, \mathbf{X}) \times$$

$$\times d\mathbf{r}_1 \, \ldots \, d\mathbf{r}_{i-1} d\mathbf{r}_{i+1} \, \ldots \, d\mathbf{r}_{j-1} d\mathbf{r}_{j+1} \, \ldots \, d\mathbf{r}_N \simeq$$

$$\simeq \overline{\rho^i_{ab}}(t, \mathbf{r}_i) \, \overline{\rho^j_{ba}}(t, \mathbf{r}_j), \tag{36}$$

along with Bogolyubov's splitting approach (see, e.g., [38]), which can describe the vanishing of the correlations between particles with time in their motion in the three-dimensional space $\mathbf{r}$:

$$\overline{f(\mathbf{r}, t) f(\mathbf{r}', t)} \simeq \overline{f(\mathbf{r}, t)} \; \overline{f(\mathbf{r}', t)}. \tag{37}$$

where

$$f(\mathbf{r}, t) = \int \sum_{i}^{N} \delta(\mathbf{r} - \mathbf{r}_i(t)) \delta(\mathbf{p} - \mathbf{p}_i(t)) d\mathbf{p} \tag{38}$$

is the space distribution function, where N is the total number of particles, and $\mathbf{p}_i(t)$ is the momentum of particle i.

At the next level of the hierarchy of approximations, we can make *hydrodynamic approximation*:
the local distribution function of particles in the six-dimensional velocity-coordinate space is the Maxwell–Boltzmann distribution describing the local equilibrium state of the atoms. Then the collisions between atoms will not contribute to the averaged evolution equation for the position of a physical volume element $d\mathbf{r}$ (Boltzmann–Enskog' integral vanishes) (see, e.g., [39]).

Therefore, we can obtain the averaged evolution equations in the following way.

The definitions of the introduced N-particle density matrix elements yield

$$\rho^i_{ab}(t, \mathbf{X}) = \left(\rho^i_{ba}(t, \mathbf{X})\right)^* ;$$

$$\rho^{i;j}_{ab;ba}(t, \mathbf{X}) = \left(\rho^{i;j}_{ba;ab}(t, \mathbf{X})\right)^* ,$$

$$\rho^{i;j}_{ba;ba}(t, \mathbf{X}) = \left(\rho^{i;j}_{ab;ab}(t, \mathbf{X})\right)^* ,$$

$$\rho^{i;j}_{aa;ba}(t, \mathbf{X}) = \left(\rho^{i;j}_{aa;ab}(t, \mathbf{X})\right)^* ,$$

$$\rho^{i;j}_{bb;ab}(t, \mathbf{X}) = \left(\rho^{i;j}_{bb;ba}(t, \mathbf{X})\right)^* , \tag{39}$$

and, correspondingly,

$$\rho^i_{ba}(t, \mathbf{X}) - \rho^i_{ab}(t, \mathbf{X}) = 2i \mathrm{Im} \left(\rho^i_{ba}(t, \mathbf{X})\right) ,$$

$$\rho^i_{ba}(t, \mathbf{X}) + \rho^i_{ab}(t, \mathbf{X}) = 2 \mathrm{Re} \left(\rho^i_{ab}(t, \mathbf{X})\right) ,$$

$$\rho^{i;j}_{ba;ba}(t, \mathbf{X}) + \rho^{i;j}_{ba;ab}(t, \mathbf{X}) - \rho^{i;j}_{ab;ab}(t, \mathbf{X}) - \rho^{i;j}_{ab;ba}(t, \mathbf{X}) =$$

$$= 2i \mathrm{Im} \left(\rho^{i;j}_{ba;ba}(t, \mathbf{X}) + \rho^{i;j}_{ba;ab}(t, \mathbf{X})\right) ,$$

$$\rho^{i;j}_{aa;ba}(t, \mathbf{X}) + \rho^{i;j}_{aa;ab}(t, \mathbf{X}) - \rho^{i;j}_{bb;ab}(t, \mathbf{X}) - \rho^{i;j}_{bb;ba}(t, \mathbf{X}) =$$

$$= 2 \, \mathrm{Re} \left(\rho^{i;j}_{aa;ba}(t, \mathbf{X}) - \rho^{i;j}_{bb;ba}(t, \mathbf{X})\right) ,$$

$$\rho^{i;j}_{ba;ba}(t, \mathbf{X}) + \rho^{i;j}_{ba;ab}(t, \mathbf{X}) + \rho^{i;j}_{ab;ab}(t, \mathbf{X}) + \rho^{i;j}_{ab;ba}(t, \mathbf{X}) =$$

$$= 2 \, \mathrm{Re} \left(\rho^{i;j}_{ba;ba}(t, \mathbf{X}) + \rho^{i;j}_{ba;ab}(t, \mathbf{X})\right) . \tag{40}$$

For convenience, we introduce the following notation:

$$P^{i;j} \left(t, \mathbf{X}\right) = \rho^{i;j}_{ba;ba}(t, \mathbf{X}) + \rho^{i;j}_{ba;ab}(t, \mathbf{X}); \tag{41}$$

and

$$R^{i;j} \left(t, \mathbf{X}\right) = \rho^{i;j}_{aa;ba}(t, \mathbf{X}) - \rho^{i;j}_{bb;ba}(t, \mathbf{X}); \tag{42}$$

After averaging each of Eqs. (5)–(8) with the distribution $\mathcal{D}(t, \mathbf{X}, \dot{\mathbf{X}})$ (the probability to find the system of atoms at the phase domain $(\mathbf{X} \pm d\mathbf{X}, \dot{\mathbf{X}} \pm d\dot{\mathbf{X}})$ in the time interval $t \pm dt$) in accordance with rule (35), we obtain the system of equations given below. For ρ^i_{aa}:

$$\int d\mathbf{r}_1 d\mathbf{r}_2 \ldots d\mathbf{r}_{i-1} d\mathbf{r}_{i+1} \ldots d\mathbf{r}_N \, D(t, \mathbf{X}) \frac{\partial}{\partial t} \rho^i_{aa}(t, \mathbf{X}) =$$

$$= \lambda_a \frac{n\left(\mathbf{r}_i\right)}{N} - \gamma \overline{\rho^i_{aa}} \left(t, \mathbf{r}_i\right) + \frac{i}{\hbar} \left(\overline{\rho^i_{ba}} \left(t, \mathbf{r}_i\right) \wp^i_{ab} \cdot \mathbf{E} \left(t, \mathbf{r}_i\right) - \right.$$

$$\left. - \overline{\rho^i_{ab}} \left(t, \mathbf{r}_i\right) \wp^i_{ba} \cdot \mathbf{E}^* \left(t, \mathbf{r}_i\right) \right) + \frac{2}{4\pi\varepsilon_0} \frac{1}{\hbar} \sum_{j;j \neq i}^{N} \int d\mathbf{r}_j \times$$

$$\times\left\{\mathrm{Im}\left(\overline{P^{i;j}}\left(t,\mathbf{r}_i,\mathbf{r}_j\right)\right)Q_{ab}^{ij}\left(\mathbf{r}_{ij}\right)\right\}; \tag{43}$$

for ρ_{bb}^i:

$$\int d\mathbf{r}_1 d\mathbf{r}_2\ldots d\mathbf{r}_{i-1}d\mathbf{r}_{i+1}\ldots d\mathbf{r}_N\, D(t,\mathbf{X})\frac{\partial}{\partial t}\rho_{bb}^i(t,\mathbf{X})=$$
$$=\lambda_b\frac{n\left(\mathbf{r}_i\right)}{N}-\gamma\overline{\rho_{bb}^i}\left(t,\mathbf{r}_i\right)-$$
$$-\frac{i}{\hbar}\left(\overline{\rho_{ba}^i}\left(t,\mathbf{r}_i\right)\wp_{ab}^i\cdot\mathbf{E}\left(t,\mathbf{r}_i\right)-\overline{\rho_{ab}^i}\left(t,\mathbf{r}_i\right)\wp_{ba}^i\cdot\mathbf{E}^*\left(t,\mathbf{r}_i\right)\right)-$$
$$-\frac{2}{4\pi\varepsilon_0}\frac{1}{\hbar}\sum_{j;j\neq i}^N\int d\mathbf{r}_j\left\{\mathrm{Im}\left(\overline{P^{i;j}}\left(t,\mathbf{r}_i,\mathbf{r}_j\right)\right)Q_{ab}^{ij}\left(\mathbf{r}_{ij}\right)\right\}; \tag{44}$$

and for ρ_{ba}^i:

$$\int d\mathbf{r}_1 d\mathbf{r}_2\ldots d\mathbf{r}_{i-1}d\mathbf{r}_{i+1}\ldots d\mathbf{r}_N\, D(t,\mathbf{X})\frac{\partial}{\partial t}\rho_{ba}^i(t,\mathbf{X})=$$
$$=-\gamma_{ba}\overline{\rho_{ba}^i}\left(t,\mathbf{r}_i\right)+i\overline{\omega}_0\overline{\rho_{ba}^i}\left(t,\mathbf{r}_i\right)+$$
$$+\frac{i}{\hbar}\left(\overline{\rho_{aa}^i}\left(t,\mathbf{r}_i\right)-\overline{\rho_{bb}^i}\left(t,\mathbf{r}_i\right)\right)\wp_{ab}^i\cdot\mathbf{E}^*\left(t,\mathbf{r}_i\right)-$$
$$-\frac{2}{4\pi\varepsilon_0}\frac{i}{\hbar}\sum_{j;j\neq i}^N\int d\mathbf{r}_j\left\{\mathrm{Re}\left(\overline{R^{i;j}}\left(t,\mathbf{r}_i,\mathbf{r}_j\right)\right)Q_{ab}^{ij}\left(\mathbf{r}_{ij}\right).\right\} \tag{45}$$

Finally, we present the quasiclassical equation of motion

$$m_i\int d\mathbf{r}_1 d\mathbf{r}_2\ldots d\mathbf{r}_{i-1}d\mathbf{r}_{i+1}\ldots d\mathbf{r}_N\, d\dot{\mathbf{r}}_1\ldots d\dot{\mathbf{r}}_N\times$$
$$\times\mathcal{D}\left(t,\mathbf{X},\dot{\mathbf{X}}\right)\ddot{\mathbf{r}}_i(t)=-\frac{\partial}{\partial\mathbf{r}_i}\left\{-\left(\overline{\rho_{ba}^i}(t,\mathbf{r}_i)\wp_{ab}^i\cdot\mathbf{E}(t,\mathbf{r}_i)+\right.\right.$$
$$\left.+\overline{\rho_{ab}^i}\left(t,\mathbf{r}_i\right)\wp_{ba}^i\cdot\mathbf{E}^*\left(t,\mathbf{r}_i\right)\right)+$$
$$\left.+\frac{2}{4\pi\varepsilon_0}\sum_{j;j\neq i}^N\int d\mathbf{r}_j\left\{\mathrm{Re}\left(\overline{P^{i;j}}\left(t,\mathbf{r}_i,\mathbf{r}_j\right)\right)Q_{ab}^{ij}\left(\mathbf{r}_{ij}\right)\right\}\right\}, \tag{46}$$

The averaged quantities $\overline{P^{i;j}}\left(t,\mathbf{r}_i,\mathbf{r}_j\right)$ and $\overline{R^{i;j}}\left(t,\mathbf{r}_i,\mathbf{r}_j\right)$ are defined by rule (36). Here, we used the identities

$$\overline{\omega}_0\overline{\rho_{\alpha\beta}^i}\left(t,\mathbf{r}_i\right)=$$
$$=\int d\mathbf{r}_1 d\mathbf{r}_2\ldots d\mathbf{r}_{i-1}d\mathbf{r}_{i+1}\ldots d\mathbf{r}_N\, d\dot{\mathbf{r}}_1\ldots d\dot{\mathbf{r}}_N\times$$
$$\times\rho_{\alpha\beta}^i\left(t,\mathbf{X}\right)\mathcal{D}(t,\mathbf{X},\dot{\mathbf{X}})\omega_0\left(\dot{\mathbf{r}}_i\right), \tag{47}$$

where $\alpha\in(a,\,b)$ and $\beta\in(a,\,b)$. We also have

$$\frac{n(\mathbf{r}_i)}{N}=\overline{\rho_{aa}^i}\left(t,\mathbf{r}_i\right)+\overline{\rho_{bb}^i}\left(t,\mathbf{r}_i\right)=$$
$$=\int d\mathbf{r}_1\ldots d\mathbf{r}_{i-1}d\mathbf{r}_{i+1}\ldots d\mathbf{r}_N\, d\dot{\mathbf{r}}_1 d\dot{\mathbf{r}}_2\ldots d\dot{\mathbf{r}}_N\times$$
$$\times\mathcal{D}(t,\mathbf{X},\dot{\mathbf{X}}), \tag{48}$$

where

$$D(t,\mathbf{X})=\int d\dot{\mathbf{r}}_1 d\dot{\mathbf{r}}_2\ldots d\dot{\mathbf{r}}_N\,\mathcal{D}(t,\mathbf{X},\dot{\mathbf{X}}). \tag{49}$$

The normalization condition reads

$$\int\limits_{V^N} d\mathbf{X}\, D(t,\mathbf{X})=\int d\mathbf{X}\, d\dot{\mathbf{X}}\,\mathcal{D}(t,\mathbf{X},\dot{\mathbf{X}})=1, \tag{50}$$

where the integration over each coordinate $\mathbf{r}_i$ $(i=1,\ldots,N)$ is within the volume V physically filled by a gas of atoms. Therefore, because of the normalization condition or, in other words, the particle number conservation in the volume V, the N-particle distributions $\mathcal{D}$ and D do not explicitly depend on time: the normalization condition is true for any moment time t. Then we have

$$\frac{\partial}{\partial t}\int\limits_{V^N} d\mathbf{X}\, D(t,\mathbf{X})=\frac{\partial}{\partial t}\int d\mathbf{X}\, d\dot{\mathbf{X}}\,\mathcal{D}(t,\mathbf{X},\dot{\mathbf{X}})=0$$
$$\Leftrightarrow\frac{\partial}{\partial t}D(t,\mathbf{X})=0. \tag{51}$$

Using the above identities and properties, we can represent the averaged partial derivative with respect to the time for the variables $\rho_{aa}^i\left(t,\mathbf{X}\right)$, $\rho_{bb}^i\left(t,\mathbf{X}\right)$, $\rho_{ba}^i\left(t,\mathbf{X}\right)$ in the following way:

$$\int d\mathbf{r}_1 d\mathbf{r}_2\ldots d\mathbf{r}_{i-1}d\mathbf{r}_{i+1}\ldots d\mathbf{r}_N\, D(t,\mathbf{X})\frac{\partial}{\partial t}\rho_{\alpha\alpha'}^i(t,\mathbf{X})=$$
$$=\frac{\partial}{\partial t}\int d\mathbf{r}_1 d\mathbf{r}_2\ldots d\mathbf{r}_{i-1}d\mathbf{r}_{i+1}\ldots d\mathbf{r}_N\, D(t,\mathbf{X})\rho_{\alpha\alpha'}^i(t,\mathbf{X})-$$
$$-\int d\mathbf{r}_1 d\mathbf{r}_2\ldots d\mathbf{r}_{i-1}d\mathbf{r}_{i+1}\ldots d\mathbf{r}_N\,\rho_{\alpha\alpha'}^i(t,\mathbf{X})\frac{\partial}{\partial t}D(t,\mathbf{X})=$$
$$=\frac{\partial}{\partial t}\overline{\rho_{\alpha\alpha'}^i}\left(t,\mathbf{r}_i\right), \tag{52}$$

where $\alpha=\{a,b\}$ and $\alpha^{'}=\{a,b\}$.

To do an approximation, we assume that two atoms become statistically uncorrelated through a quite small time interval such that the following expressions are true on the appropriately chosen time-scale:

$$\overline{\rho^{i;j}_{ba;ba}}\,(t,\mathbf{r}_i,\mathbf{r}_j) \approx \overline{\rho^{i}_{ba}}\,(t,\mathbf{r}_i)\;\;\overline{\rho^{j}_{ba}}\,(t,\mathbf{r}_j)\,;$$

$$\overline{\rho^{i;j}_{ba;ab}}\,(t,\mathbf{r}_i,\mathbf{r}_j) \approx \overline{\rho^{i}_{ba}}\,(t,\mathbf{r}_i)\;\;\overline{\rho^{j}_{ab}}\,(t,\mathbf{r}_j)\,;$$

$$\overline{\rho^{i;j}_{aa;ba}}\,(t,\mathbf{r}_i,\mathbf{r}_j) \approx \overline{\rho^{i}_{aa}}\,(t,\mathbf{r}_i)\;\;\overline{\rho^{j}_{ba}}\,(t,\mathbf{r}_j)\,;$$

$$\overline{\rho^{i;j}_{bb;ba}}\,(t,\mathbf{r}_i,\mathbf{r}_j) \approx \overline{\rho^{i}_{bb}}\,(t,\mathbf{r}_i)\;\;\overline{\rho^{j}_{ba}}\,(t,\mathbf{r}_j)\,. \qquad (53)$$

We describe the system that consists of only identical atoms. So, we can approximate the averaged distributions by using the symmetry properties:

$$\overline{\rho^{i}_{ba}}\,(t,\mathbf{r}_i) = \rho_{ba}\,(t,\mathbf{r})\,,$$

$$\overline{\rho^{i}_{aa}}\,(t,\mathbf{r}_i) = \rho_{aa}\,(t,\mathbf{r})\,,$$

$$\overline{\rho^{i}_{bb}}\,(t,\mathbf{r}_i) = \rho_{bb}\,(t,\mathbf{r})\,. \qquad (54)$$

Then the expressions under the sign of integration in the round brackets in the evolution equations can be presented as follows:

$$\operatorname{Im}\left(\overline{\rho^{i;j}_{ba;ba}}\,(t,\mathbf{r}_i,\mathbf{r}_j) + \overline{\rho^{i;j}_{ba;ab}}\,(t,\mathbf{r}_i,\mathbf{r}_j)\right) \approx$$

$$\approx 2\operatorname{Im}\left(\rho_{ba}\,(t,\mathbf{r})\right)\operatorname{Re}\left(\rho_{ba}\,(t,\mathbf{r}')\right);$$

$$\operatorname{Re}\left(\overline{\rho^{i;j}_{aa;ba}}\,(t,\mathbf{r}_i,\mathbf{r}_j) - \overline{\rho^{i;j}_{bb;ba}}\,(t,\mathbf{r}_i,\mathbf{r}_j)\right) \approx$$

$$\approx \left(\rho_{aa}\,(t,\mathbf{r}) - \rho_{bb}\,(t,\mathbf{r})\right)\operatorname{Re}\left(\rho_{ba}\,(t,\mathbf{r}')\right);$$

$$\operatorname{Re}\left(\overline{\rho^{i;j}_{ba;ba}}\,(t,\mathbf{r}_i,\mathbf{r}_j) + \overline{\rho^{i;j}_{ba;ab}}\,(t,\mathbf{r}_i,\mathbf{r}_j)\right) \approx$$

$$\approx 2\operatorname{Re}\left(\rho_{ba}\,(t,\mathbf{r})\right)\operatorname{Re}\left(\rho_{ba}\,(t,\mathbf{r}')\right). \qquad (55)$$

Therefore, the above system of evolution equations for the densities of atomic states becomes soluble and closed in the variables that describe only "one-particle" distributions ($\rho_{aa}\,(t,\mathbf{r})$, $\rho_{bb}\,(t,\mathbf{r})$, $\rho_{ba}\,(t,\mathbf{r})$). Furthermore, we suppose the local equilibrium distribution $\mathcal{D}$ within the volume equal to the cube of a half-wavelength $\frac{(\pi)^3}{k^3}$. Then the collision effects vanish after the averaging over the i-th particle acceleration $\frac{d\dot{\mathbf{r}}_i(t)}{dt}$:

$$\int d\mathbf{r}_1 \ldots d\mathbf{r}_{i-1} d\mathbf{r}_{i+1} \ldots d\mathbf{r}_N\, d\dot{\mathbf{r}}_1 d\dot{\mathbf{r}}_2 \ldots d\dot{\mathbf{r}}_N\, \mathcal{D}\times$$

$$\times(t,\mathbf{X},\dot{\mathbf{X}})\ddot{\mathbf{r}}_i(t) = \frac{d}{dt}\int d\mathbf{r}_1 d\mathbf{r}_2 \ldots d\mathbf{r}_{i-1} d\mathbf{r}_{i+1} \ldots d\mathbf{r}_N\times$$

$$\times d\dot{\mathbf{r}}_1 \ldots d\dot{\mathbf{r}}_N \mathcal{D}(t,\mathbf{X},\dot{\mathbf{X}})\dot{\mathbf{r}}_i(t) - \int d\mathbf{r}_1 d\mathbf{r}_2 \ldots d\mathbf{r}_{i-1}\times$$

$$\times d\mathbf{r}_{i+1} \ldots d\mathbf{r}_N\; d\dot{\mathbf{r}}_1 d\dot{\mathbf{r}}_2 \ldots d\dot{\mathbf{r}}_N\; \dot{\mathbf{r}}_i \frac{d}{dt}\mathcal{D}(t,\mathbf{X},\dot{\mathbf{X}}) \approx$$

$$\approx \frac{d}{dt}\mathbf{u}\,(t,\mathbf{r}) = \frac{\partial}{\partial t}\mathbf{u}\,(t,\mathbf{r}) + \left(\mathbf{u}\,(t,\mathbf{r})\cdot\frac{\partial}{\partial \mathbf{r}}\right)\mathbf{u}\,(t,\mathbf{r})\,, \qquad (56)$$

where

$$\frac{d}{dt}\mathcal{D}(t,\mathbf{X},\dot{\mathbf{X}}) = \frac{\partial}{\partial \mathbf{X}}\mathcal{D}(\mathbf{X},\dot{\mathbf{X}})\frac{d}{dt}\mathbf{X} + \frac{\partial}{\partial \dot{\mathbf{X}}}\mathcal{D}(\mathbf{X},\dot{\mathbf{X}})\times$$

$$\times\frac{d^2}{dt^2}\mathbf{X} = -\frac{\mathcal{D}(\mathbf{X},\dot{\mathbf{X}})}{J(t,\mathbf{X},\dot{\mathbf{X}})}\left[\frac{\partial}{\partial t}J(t,\mathbf{X},\dot{\mathbf{X}}) + \frac{\partial}{\partial \mathbf{X}}J\times\right.$$

$$\left.\times(t,\mathbf{X},\dot{\mathbf{X}})\cdot\dot{\mathbf{X}} + \frac{\partial}{\partial \dot{\mathbf{X}}}J(t,\mathbf{X},\dot{\mathbf{X}})\cdot\frac{d^2}{dt^2}\mathbf{X}\right]. \qquad (57)$$

Here, $J(t,\mathbf{X},\dot{\mathbf{X}})$ is the Jacobian (determinant of the Jacobian matrix $\hat{J}$) of the transformation of phase coordinates along the phase trajectories with time for the atoms:

$$\left(\mathbf{X}(t+dt),\dot{\mathbf{X}}(t+dt)\right) = \hat{J}\cdot\left(\mathbf{X}(t),\dot{\mathbf{X}}(t)\right). \qquad (58)$$

Assuming $\wp^i_{ab} = \wp^i_{ba} = \wp\hat{\wp}$, $i = 1,\ldots,N$, and distinguishing the directions parallel or antiparallel to the external field $\mathbf{E}$, we obtain the following system of the evolution equations.
For $\rho_{aa}\,(t,\mathbf{r})$:

$$\frac{\partial}{\partial t}\rho_{aa}\,(t,\mathbf{r}) = \lambda_a\frac{n\,(\mathbf{r})}{N} - \gamma\rho_{aa}\,(t,\mathbf{r}) +$$

$$+\frac{i}{\hbar}\wp\left(\rho_{ba}\,(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}\,(t,\mathbf{r}) - \rho_{ab}\,(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}^*\,(t,\mathbf{r})\right) +$$

$$+\frac{2}{\hbar}\chi\int d\mathbf{r}'\{2\operatorname{Im}\left(\rho_{ba}(t,\mathbf{r})\right)\operatorname{Re}\left(\rho_{ba}(t,\mathbf{r}')\right)Q\,(\mathbf{r},\mathbf{r}')\}; \qquad (59)$$

for $\rho_{bb}\,(t,\mathbf{r})$:

$$\frac{\partial}{\partial t}\rho_{bb}\,(t,\mathbf{r}) = \lambda_b\frac{n\,(\mathbf{r})}{N} - \gamma\rho_{bb}\,(t,\mathbf{r}) -$$

$$-\frac{i}{\hbar}\wp\left(\rho_{ba}\,(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}\,(t,\mathbf{r}) - \rho_{ab}\,(t,\mathbf{r})\,\hat{\wp}\cdot\mathbf{E}^*\,(t,\mathbf{r})\right) -$$

$$-\frac{2}{\hbar}\chi\int d\mathbf{r}'\{2\operatorname{Im}\left(\rho_{ba}(t,\mathbf{r})\right)\operatorname{Re}\left(\rho_{ba}(t,\mathbf{r}')\right)Q\,(\mathbf{r},\mathbf{r}')\}; \qquad (60)$$

for $\rho_{ba}\left(t,\mathbf{r}\right)$:

$$\frac{\partial}{\partial t}\rho_{ba}(t,\mathbf{r}) = -\gamma_{\mathbf{ba}}\rho_{\mathbf{ba}}(\mathbf{t},\mathbf{r}) + \mathbf{i}\overline{\omega}_{\mathbf{0}}\rho_{\mathbf{ba}}(\mathbf{t},\mathbf{r}) +$$

$$+\frac{i}{\hbar}(\rho_{aa}(t,\mathbf{r}) - \rho_{\mathbf{bb}}(\mathbf{t},\mathbf{r}))\wp\hat{\wp}\cdot\mathbf{E}^{*}(\mathbf{t},\mathbf{r}) -$$

$$\frac{2i}{\hbar}\chi\int d\mathbf{r}'\{(\rho_{aa}(t,\mathbf{r}) - \rho_{bb}(t,\mathbf{rt})\mathrm{Re}(\rho_{\mathbf{ba}}(\mathbf{t},\mathbf{r}'))\mathbf{Q}(\mathbf{r},\mathbf{r}')\}. \tag{61}$$

Finally, we present the equation for the translational motion of a quasiclassical particle:

$$m\frac{\partial}{\partial t}\mathbf{u}(t,\mathbf{r}) = -m\left(\mathbf{u}(t,\mathbf{r})\cdot\frac{\partial}{\partial\mathbf{r}}\right)\mathbf{u}(t,\mathbf{r}) -$$

$$-\frac{\partial}{\partial\mathbf{r}}\left\{-\wp\left(\rho_{ba}(t,\mathbf{r})\hat{\wp}\cdot\mathbf{E}(t,\mathbf{r}) + \rho_{ab}(t,\mathbf{r})\hat{\wp}\cdot\mathbf{E}^{*}(t,\mathbf{r})\right) +\right.$$

$$\left.+2\chi\int d\mathbf{r}'\left\{2\mathrm{Re}\left(\rho_{ba}\left(t,\mathbf{r}\right)\right)\mathrm{Re}\left(\rho_{ba}\left(t,\mathbf{r}'\right)\right)Q\left(\mathbf{r},\mathbf{r}'\right)\right\}\right\}. \tag{62}$$

This proves the validity of (12)–(15).

4. Conclusion

Thus, we have constructed the microscopic and macroscopic kinetic equations for the density matrix elements of N-atom states including the atomic motion with regard for the atom-field and atom-atom interactions.

This theory with a certain modification can be applied to the description of the phenomena involving collective effects. The approximation used here can be related to the systems of quite "hot" and dense atomic gases interacting with the parallel or antiparallel near-resonant laser beams. Such systems were investigated experimentally (see, e.g., [3, 6]). The main difference between the approach in [27] and our one lies in that the results in [27] are applicable to the quite "cold" atomic gases with the restrictions mentioned above and to the radiation processes rather "slow" in comparison with the spontaneous radiation time of atoms (as in [23, 30]). As a possible application of the theory, the analytical solutions can be obtained for the resulting macroscopic equations in different limits, for example, for a system composed of a strong coherent "pumping" field and a weak counter-propagating "probe" field.

Another interesting "quite" fundamental result deserves a few words here. As we have seen, the used model Hamiltonian allows one to model the dipole-dipole interaction between atoms and to build in a the microscopic kinetic equations for the density matrix elements of a system in a straightforward manner. Other methods involving Green's functions [13–17] can be more cumbersome and, generally saying, do not allow one to formulate the kinetic equations (including the short-range and long-range "collision integrals") in terms of the convenient density matrix elements.

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ДИНАМІЧНИЙ ТА МАКРОСКОПІЧНИЙ ОПИСИ ГАЗУ ВЗАЄМОДІЮЧИХ АТОМІВ У СИЛЬНОМУ ЕЛЕКТРОМАГНІТНОМУ ПОЛІ ПОБЛИЗУ РЕЗОНАНСУ

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Резюме

Робота присвячена побудові мікроскопічної та макроскопічної теорій системи N взаємодіючих дворівневих атомів у сильному та слабкому електромагнітних полях. Побудовані мікроскопічні кінетичні рівняння для матричних елементів густини станів атомів та атомного руху N-атомної системи, які враховують взаємодію між атомами та атомів із полем. Відповідна макроскопічна кінетика побудована для одно- та двочастинкової функцій розподілів матриці густини атомних станів. Самоузгоджена система макроскопічних одночастинкових рівнянь для усереднених елементів матриці густини атомних станів разом із рівняннями Максвелла дозволяє описувати випромінювальні та поглинальні властивості системи і пояснити залежність оптичних властивостей від густини частинок у термінах далекодійної диполь-дипольної взаємодії між атомами.