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INTRODUCING THE GENERALIZED ABSORPTANCE FOR A GAS WITH BOUND ATOMIC STATES

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The quantum optical theory of absorption/reemission properties of a system of interacting atoms is discussed. The calculation method of the absorption coefficient is developed with regard for the quantization of field, thermal atomic motion, Doppler effect, and the model interaction between atoms. It is shown that the formulation of the absorption coefficient in the quantum optical context is based on the commutation relation between the operators of electric field and intensity. The revealed non-linear dependence of the absorption coefficient on the atomic density, even in the case of negligible binary interaction, can be referred to a certain kind of quantum-optic collective effects.

K e y w o r d s: absorption coefficient, quantum optics, Doppler effect, commutation relation, many-body interaction.

1. Introduction

In this paper, the definition and methodology of numerical evaluation of the absorption coefficient is presented for a system of interacting two-level atoms coupled with a quantized electromagnetic field. The presented formalism is based on a model Hamiltonian. In comparison with other models (e.g., used in such works as [1]–[4]), we explicitly take the atomic motion in space into consideration within the model Hamiltonian, and the induced Doppler effect is naturally introduced into the corresponding averaging procedure. The local absorptance is defined as the intensity logarithm derivative with respect to the distance in a vicinity of the given spatial point and the moment of time. The intensity is represented by the averaged normally ordered product of the creation and annihilation operators of certain states of the electromagnetic field. The averaging is performed over all possible quantum states of the total system of the field and atoms. Approximating the model of the system by the time-independent Hamiltonian, the general expression for the intensity is expanded into the series in terms of the commutation relations between the Hamiltonian and the intensity operator. In comparison with the method of the impact theory, the chronological order of events in the system is not discussed here (compare, e.g., with [5]). Under certain deterministic assumptions for the system placed into a perfect cavity, the formulation of the Maxwell–Bloch equations (or the so-called optical Bloch equations, see [6]) describing the evolution of the system in terms of one-particle population/polarization matrix elements is not discussed.

As shown in the second part of this research, the assumption about some initial thermodynamic equilibrium corresponding to the average over a statistical ensemble amends the properties of the system as a whole, which is of certain interest. Namely, the ensemble-averaged value of a physical quantity can be found complex, as it will be shown elsewhere. In

comparison with the commonly used introduction of an explicit phenomenological non-Hermitian item (see, e.g., [7]) into the model Hamiltonian describing the decay phenomena and therefore defining a finite absorption line width in the dipole approximation, the source of non-ideality is expected here from the model statistical distribution of atomic states. In the used assumptions, the decay of atomic states can be implicitly included in the dynamics of the system. Furthermore, some non-ideality is imposed by the defined averaging procedure (see the discussion, e.g., in [8] and [9]). In other words, the introduction of the concept of measuring the intensity "takes off" the thermodynamic isolation (e.g., the particle conservation) and, generally speaking, requires the consideration of certain dissipative processes. Technically, we avoid the explicit formulation of a damping operator in our discussion. The detailed description of the used model is provided below.

Furthermore, the model introduced here also involves the collective interaction between atoms induced by the mediatorial electromagnetic field. The proposed method potentially allows one to account for collective level shifts and the line broadening arising from the photon exchange between atoms and being observed experimentally ([10]–[15]). The theory can be useful in the applications to many-body quantum science.

2. Model of the System

Assume that the system of N atoms occupies the volume V bounded by optically transparent and thermally insulated (e.g., up to the near-infrared region) walls. Suppose also that the electromagnetic field generated within a certain period of time "reaches" the space of volume $V_{\rm el}$ (which includes V). Atoms that fill V are able to interact with the modes of this electromagnetic field. The frequencies of the modes are distributed near the resonant frequency ω_0 of a single atomic (or molecular) transition between only two levels b (ground state) and a (excited state). The modes of the electromagnetic field correspond to Fock states (or the number states with a well-defined number of quanta) formally obtained through the standard procedure of quantization in the volume $V_{\rm el}$. Moreover, the atoms interacting on relatively short distances (e.g., of the order of the van der Waals radius) by means of a potential U are able to absorb or emit quanta of the electromagnetic field in some high-frequency domain (in the optical region). The interaction of the field with an atom is determined through the scalar potential formed by the scalar (inner) product of the atomic dipole moment (more carefully, the dipole matrix) and the operator of electric field. The translational motion of each atom has three degrees of freedom and is confined by rigid walls (forming the volume V). Therefore, the infinitesimal change of the state function of the system during the infinitesimal time intervals is described by the following Hamiltonian:

$$\hat{\mathcal{H}} = \sum_{i=1}^{N}\left(\frac{\hat{\mathbf{p}}_i^2}{2M_i} + \hat{\mathcal{H}}_i^{\rm rel} + \sum_{j=1;i<j}^{N} U_{ij} - \hat{\mathbf{d}}_i\cdot\mathbf{E}(\mathbf{r}_i)\right) + \hat{\mathcal{H}}^F. \quad (1)$$

Here, the first two terms describe the spatial "movement" of N atoms with the momenta $\hat{\mathbf{p}}_i$ and intrinsic energies $\hat{\mathcal{H}}_i^{\rm rel}$ (for $i = 1, ..., N$). M_i denotes the i-th atomic mass. The third term ($U_{ij} \equiv U\left(r_{ij}, \alpha_i, \alpha_j\right)$) gives the potential energy of all pairs of atoms depending on the interatomic distances $r_{ij} \equiv |\mathbf{r}_{ij}| = |\mathbf{r}_i - \mathbf{r}_j|$ and the internal atomic states α_i, α_j. The fourth term is the energy of interaction between the atoms and the electromagnetic field $\mathbf{E}$ expressed by the standard dipole-field coupling. Here, in the coordinate representation, the dipole moment operator for the i-th atom is defined as $\hat{\mathbf{d}}_i = \sum_{v=1}^{\rm Val}\left(-e\hat{\mathbf{r}}_{\rm iv}^{\rm rel}\right)$ taking all the Val valent (unpaired) electrons into account, e is the absolute value of electron charge, $\hat{\mathbf{r}}_{\rm iv}^{\rm rel}$ is the position of the v-th valent electron relative to the nucleus of i-th atom, and $\hat{\mathcal{H}}^F$ represents the energy of the free electromagnetic field in the volume $V_{\rm el}$.

If the bound states between any two atoms in the system are taken into account, then, in the coordinate representation, we expand the state if the system in the basis vectors of the Hilbert space defined as the product of the subspaces of the eigenvectors $|\mathbf{r}_i\rangle$ and $|\alpha_i\rangle$, $|\mathbf{r}_d\rangle$ and $|\mathbf{r}_{ij}\rangle$ and $|\alpha_d, \alpha_d'\rangle$ for $i = 1, ..., N$ and $d = 1, ..., D$. The eigenvectors from the subspaces are, by definition, the solutions of the corresponding stationary Schrödinger equations. Therefore, denoting $|\varepsilon, \mathbf{r}, \alpha\rangle_i \equiv |i\rangle$, it satisfies the eigenvalue equation such as $\hat{\mathcal{H}}_i^A\,|i\rangle = (\varepsilon_i + \hbar\omega_{\alpha_i})\,|i\rangle$, giving the sum of translational kinetic and intrinsic energies for a free atom indexed as the i-th one. Here, the quantum of energy $\hbar\omega_{\alpha_i}$ equals the energy of a valent electron

(electrons) on the energy level indexed by the symbol α_i, $i = 1, ..., N$.

The subspace of bound (weakly bound) two-atomic states consists of such subspaces as $|\mathbf{r}_d\rangle$ defining the translational motion of the center of masses for the coupled i-th and j-th atoms, $|\mathbf{r}_{ij}\rangle$ defining the motion of atoms relative to the center of masses in a bound state (such as rotation and vibration), and the state of the valent electrons in the pair of atoms denoted by $|\alpha_d, \alpha'_d\rangle$. Formally, symbolizing $|\mathbf{r}_d\rangle\ |\mathbf{r}_{ij}\rangle\ |\alpha_d, \alpha'_d\rangle$ as the vector $|d\rangle$, we have $\left(\hat{\mathcal{H}}_i^A + \hat{\mathcal{H}}_j^A + U_{ij}\right)|d\rangle = (\varepsilon_d + E_d + \hbar\omega_\alpha + \hbar\omega_{\alpha'})\,|d\rangle$ giving the sum of the translational $\frac{p_d^2}{2M_d}$, rotational and vibrational E_d, and "intrinsic" electronic $\hbar\omega_\alpha + \hbar\omega_{\alpha'}$ energies for a coupled two-atomic system indexed by d.

As was noted above, the electromagnetic field can be defined through the superposition of its modes determined by the corresponding Fock states. Then the total "free" field energy can be represented through the sum of the corresponding quanta of energy: $\hat{\mathcal{H}}^F = \sum_q \hat{\mathcal{H}}_q^F$, where the symbol q denotes the mode of the electromagnetic field with the propagation wave vector $\mathbf{k}$ and the j-th polarization with $j = 1, 2$ (see details, e.g., in [16]). Accordingly, the Fock states denoted by $|n_q\rangle$ are the eigenvectors of the corresponding operators $\hat{\mathcal{H}}_q^F|n_q\rangle = \left(n_q + \frac{1}{2}\right)\hbar\omega_q|n_q\rangle$. Recall that the state $|n_q\rangle$ determines the number of photons n corresponding to the mode q.

Therefore, we can introduce the pure quantum state of the system as the superposition

$$|\Psi\rangle = \sum_{\{\,\}} C_{\{\,\}}(t) \prod_{i_\ell=1}^{N-2D} |i_\ell\rangle \prod_{d_\ell=1}^{D} |d_\ell\rangle \prod_q |n_q\rangle, \qquad (2)$$

where

$$q \equiv (\mathbf{k}, j), \text{ and } \{\,\} \equiv \ell, \{\varepsilon\}, D, \{\alpha\}, \{\alpha_d\alpha'_d\}, \{n_q\}.$$

The all possible permutations of the atoms between the unbound and bound states are indexed by the symbol ℓ. Note that the number of the Fock states (photons) indexed by n_q is not limited from above.

Let us define the following unit operator:

$$\hat{1} = \sum_{\{\,\}} \prod_{i_\ell=1}^{N-2D} \prod_{d_\ell=1}^{D} \prod_q |n_q\rangle|d_\ell\rangle|i_\ell\rangle\langle i_\ell|\langle d_\ell|\langle n_q|. \qquad (3)$$

The corresponding representation of the initial Hamiltonian can be derived, by multiplying each of the terms in (1) by the unit operator $\hat{1}$ from the left- and the right-hand sides. Then, assuming the normalization and completeness of the defined subspaces, the total Hamiltonian (1) in the introduced basis (2) can be factorized in the following way:

$$\hat{\mathcal{H}} \to \hat{\mathcal{H}}'^A + \hat{\mathcal{H}}^D + \hat{\mathcal{H}}^F + \hat{\mathcal{H}}'^{AA} + \\ + \hat{\mathcal{H}}^{AD} + \hat{\mathcal{H}}'^{AF} + \hat{\mathcal{H}}^{DD} + \hat{\mathcal{H}}^{DF}. \qquad (4)$$

By definition, $\hat{\mathcal{H}}'^A \equiv \sum_{\ell,D}\sum_{i_\ell=1}^{N-2D} \hat{\mathcal{H}}_{i_\ell}^A$, where the notation D means the total number of bound states for the given permutation ℓ. The maximum amount of the coupled states is $N/2$ for an even number of atoms, while the total number of available permutations of the pair of indices without repeating is $N(N-1)/2$. $\hat{\mathcal{H}}_{i_\ell}^A \equiv Tr_{d_\ell,q}\hat{1}\hat{\mathcal{H}}_i^A\hat{1}$. We have also $\hat{\mathcal{H}}^D \equiv \sum_{\ell,D}\sum_{d_\ell=1}^{D} \hat{\mathcal{H}}_{d_\ell}^A$, where, by analogy, $\hat{\mathcal{H}}_{d_\ell}^A \equiv \equiv Tr_{i_\ell,q}\hat{1}\hat{\mathcal{H}}_i^A\hat{1}$. The symbol $Tr_{d_\ell,q}$ means that only the uncoupled (unbound) states of atoms held in the summation over all the permutations $\{\ell\}$, while $Tr_{i_\ell,q}$ means that only the coupled (bound) states of atoms held in the summation over all the permutations $\{\ell\}$. We have also

$$\hat{\mathcal{H}}'^{AF} \equiv Tr_{d_\ell,q}\,\hat{1}\,\hat{\mathcal{H}}^{AF}\hat{1} = \sum_{\ell,D}\sum_{i_\ell=1}^{N-2D} \mathcal{F}\hat{\mathbf{S}}_{i_\ell}\cdot\mathcal{E}(\mathbf{r}_{i_\ell}), \quad (5)$$

where the notation $\mathcal{F}$ imposes special additional limitations on the terms under the sign of sum in (5). Mainly, this introduces the corrections in accordance with the laws of conservations (total momentum and energy conservation during We have the scattering of a photon on a single atom) and the relativistic Doppler effect influencing the resulting absorption or emission frequencies. Therefore, for nonrelativistic atomic velocities $\sqrt{2\varepsilon_i/M_i} \ll c$, where c equals the speed of light in vacuum, the following restrictions on the acts of photon absorption or emission composing the recoil of a single atom should be noted:

$$\begin{aligned} &\mathbf{p}_i \pm \hbar\mathbf{k} = \mathbf{p}'_i,\ +(-)\ \ \text{signforabsorption(emission)};\\ &\varepsilon_i + \hbar\nu_k = \varepsilon'_i + \hbar\omega_0^{\text{rel}}\left(\mathbf{p}_i, \hat{k}\right),\\ &\omega_0^{\text{rel}} \simeq \left(1 + \frac{\hat{k}\cdot\mathbf{p}_i}{M_i\,c}\right)\omega_0. \end{aligned} \qquad (6)$$

Here, $\frac{\hat{k}\cdot\mathbf{p}_i}{M_i\,c} = -\frac{v_i}{c}\cos\theta_s$, $\hat{k} = \frac{\mathbf{k}}{|\mathbf{k}|}$ is the unit vector directed along the wave vector $\mathbf{k}$, and θ_s is the angle

between the atomic velocity of the i-th atom v_i and the direction to the (here imaginable) source of the photon at the point of absorption (see, e.g., [17]). By analogy, we have

$$\hat{\mathcal{H}}^{DF} \equiv Tr_{i_\ell,q}\hat{1}\hat{\mathcal{H}}^{AF}\hat{1} = \sum_{\ell,D}\sum_{d_\ell=1}^{D}\mathcal{F}\hat{\mathbf{S}}_{d_\ell}\cdot\mathcal{E}\left(\mathbf{r}_{d_\ell}\right) \qquad (7)$$

with the applied restriction of the laws of conservations and the Doppler frequency shift:

$$\begin{aligned}
&\mathbf{p}_d \pm \hbar\mathbf{k} = \mathbf{p}'_d,\ +(-)\ \ \text{signforabsorption(emission)};\\
&\varepsilon_d + E_d + \hbar\nu_k = \varepsilon'_d + E'_d + \hbar\omega_0^{\text{rel}}\left(\mathbf{p}_d,\hat{k}\right),\\
&\omega_0^{\text{rel}} \simeq \left(1 + \frac{\hat{k}\cdot\left(\mathbf{p}_d + \mathbf{p}_d^{\text{rel}}\right)}{M_{i,d}\,c}\right)\omega_0,
\end{aligned} \qquad (8)$$

where, within the classical interpretation, $\mathbf{p}_d^{\text{rel}}$ means the relative impulse (momentum) in the two-body problem corresponding to the motion of the i-th atom (molecule) relative to the center of masses of the d-th complex.

In a similar manner, projecting the shot-range interaction energy $\hat{\mathcal{H}}^{AA}$ onto the defined basis of states, we can split the corresponding sum over all permutations $\{\ell\}$ into the term "containing" only unbound states, and corresponding to the interaction between "free" and coupled atoms, and the interaction between the pairs of bound atoms (in bound states). For example,

$$\hat{\mathcal{H}'}^{AA} \equiv \sum_{\ell,D,n_q}\sum_{i_\ell<j_\ell}^{N-2D,N-2D}\sum_{m=0}^{n}\left[\widehat{AA}_m + Adj.\right], \qquad (9)$$

where $n = 2$, $\widehat{AA}_m = \mathcal{F}\hat{U'}_m\left(\mathbf{r}_{i_\ell j_\ell}, n_q\right)|n_q - m\rangle\langle n_q|$. Here, $\hat{U'}_m\left(\mathbf{r}_{i_\ell j_\ell}, n_q\right)$ is the operator consisting of the products of matrix elements of the potential energy U and projection operators on the unbound states from the basis in (3). Then, by analogy, we can write the short-range term for the atom-dimer, $\hat{\mathcal{H}}^{AD}$, and for dimer-dimer, $\hat{\mathcal{H}}^{DD}$, interactions. But now, instead of $\hat{U'}_m$, we can use the notation $\hat{U}_m^{(d)}\left(\mathbf{r}_{i_\ell d_\varsigma}, n_q\right)$ for the first type of interaction and $\hat{U}_m^{(dd)}\left(\mathbf{r}_{d_\ell d_\varsigma}, n_q\right)$ for the second type, accordingly. In addition, we have $n = 3$ in the first case of atom-dimer coupling and $n = 4$ in the second case, $\mathbf{r}_{i_\ell d_\varsigma}$ is the distance between the i_ℓ-th atom and the center of masses of the d_ς-th pair of atoms, and $\mathbf{r}_{d_\ell d_\varsigma}$ is the distance between the centers of masses of the d_ℓ-th and d_ς-th pairs of atoms.

In the case of the Hamiltonian independent of time, the state function (2) can formally be represented in the form

$$|\Psi\rangle\,(t) = e^{-\frac{i}{\hbar}\hat{\mathcal{H}}t}|\Psi\rangle_0, \qquad (10)$$

where $|\Psi\rangle_0$ stands for the value of the state function at the initial moment of time.

Then, in accordance with the accepted model of a device counting photons (see, e.g., [16] and [18]), the local absorption coefficient is defined as follows:

$$\alpha_{\text{tot}} \approx \frac{\delta}{\delta z}\ln\sum_{\Psi}\langle\Psi|_0\hat{\mathcal{I}}_\tau\hat{\rho}_\Psi|\Psi\rangle_0, \qquad (11)$$

where

$$\hat{\mathcal{I}}_\tau \approx \hat{\mathcal{I}} + \frac{i}{\hbar}\tau\left[\hat{\mathcal{H}},\hat{\mathcal{I}}\right] + \frac{1}{2}\left(\frac{i}{\hbar}\tau\right)^2\left[\hat{\mathcal{H}},\left[\hat{\mathcal{H}},\hat{\mathcal{I}}\right]\right],$$

and

$$\hat{\mathcal{I}} = \mathcal{E}_+\mathcal{E}_-,$$

with

$$\mathcal{E}_+\left(\mathbf{r}\right) = \sum_q \hat{e}_q\mathcal{E}_q e^{-i\mathbf{k}_q\cdot\mathbf{r}}a_q^\dagger,$$

and

$$\mathcal{E}_-\left(\mathbf{r}\right) = \sum_{q'} \hat{e}_{q'}\mathcal{E}_{q'} e^{i\mathbf{k}_{q'}\cdot\mathbf{r}}a_{q'}.$$

The statistical distribution over the pure states $|\Psi\rangle$ of the system at the initial moment of time $t = 0$ is given by the operator $\hat{\rho}_\Psi$. It was assumed that the optical pathway inside the sample is parallel to the Z-axis. Therefore, $r = z$. The phase factor in the mode amplitude $\mathcal{E}_q = \left(\frac{\hbar\nu}{2\varepsilon_0 V_{\text{el}}}\right)^{1/2}$ is neglected for simplicity. Note that the electric field operator can be represented as the sum of its constituents $\mathcal{E}_+\left(\mathbf{r}\right)$ and $\mathcal{E}_-\left(\mathbf{r}\right)$.

3. Calculation of the Commutators

We are interested in the calculation of such commutators as $\left[\hat{\mathcal{H}},\hat{\mathcal{I}}\right]$, $\left[\hat{\mathcal{H}},\left[\hat{\mathcal{H}},\hat{\mathcal{I}}\right]\right]$, ... Based on the commutation relations for the boson field operators $\left[a_q, a_{q'}^\dagger\right] = \delta_{q\,q'}$, we obtain the expression

$$\left[\hat{\mathcal{H}'}^{AF},\hat{\mathcal{I}}\right] = \sum_{\ell,D}\sum_{i_\ell=1}^{N-2D}\mathcal{F}\hat{\mathbf{S}}_{i_\ell}\cdot\left[\hat{\mathcal{E}}(\mathbf{r}_{i_\ell}),\hat{\mathcal{I}}\left(\mathbf{r}\right)\right], \qquad (12)$$

where

$$\left[\hat{\mathcal{E}}\left(\mathbf{r}_i\right),\hat{\mathcal{I}}\left(\mathbf{r}\right)\right]=\sum_{q,\tilde{q}}\hat{e}_q\mathcal{E}_q^2\left(\hat{e}_q\cdot\hat{e}_{\tilde{q}}\right)\mathcal{E}_{\tilde{q}}\times$$
$$\times\left(e^{i(\mathbf{k}_{\tilde{q}}-\mathbf{k}_q)\cdot\mathbf{r}}e^{i\mathbf{k}_q\cdot\mathbf{r}_i}a_{\tilde{q}}-Adj.\right). \quad (13)$$

Using the defined action of the photon creation and annihilation operators on the Fock states such as $a_{q'}^{\dagger}|n_q\rangle=\sqrt{n+1}|n_q+1\rangle$ and $a_{q'}|n_q\rangle=\sqrt{n}|n_q-1\rangle$, the following notations can be held in the Hilbert space:

$$\widehat{\mathcal{AA}}_{m,n_q}^{q}\left(\mathbf{r}_{ij}\right)=\mathcal{F}\hat{U'}_m\left(\mathbf{r}_{ij},n_q\right)|n_q-m\rangle\langle n_q|-Adj., \quad (14)$$

$$\widehat{\mathcal{AA}}_{m,n_q}^{q,\,q',-}\left(\mathbf{r}_{ij}\right)=$$
$$=\mathcal{F}\hat{U'}_m\left(\mathbf{r}_{ij},n_q\right)\left\{\sqrt{n_q}|n_q-m\rangle\langle n_q-1|a_{\tilde{q}'}-\right.$$
$$\left.-a_{\tilde{q}'}\sqrt{n_q-m+1}|n_q-m+1\rangle\langle n_q|\right\}+$$
$$+\mathcal{F}\overline{\hat{U}'}_m\left(\mathbf{r}_{ij},n_q\right)\left\{\sqrt{n_q-m}|n_q\rangle\langle n_q-m-1|a_{\tilde{q}'}-\right.$$
$$\left.-a_{\tilde{q}'}\sqrt{n_q+1}|n_q+1\rangle\langle n_q-m|\right\}, \quad (15)$$

$$\widehat{\mathcal{AA}}_{m,n_q}^{q,\,q',+}\left(\mathbf{r}_{ij}\right)=-\left(\widehat{\mathcal{AA}}_{m,n_q}^{q,\,q',-}\left(\mathbf{r}_{ij}\right)\right)^{\dagger}. \quad (16)$$

Therefore, the commutators of interatomic interaction operators with the intensity operator can be written as follows:

$$\left[\hat{\mathcal{H}}_{ij}^{'AA},\hat{\mathcal{I}}\left(\mathbf{r}\right)\right]=$$
$$=\sum_m\sum_q\left\{m\,\mathcal{E}_q^2\widehat{\mathcal{AA}}_{m,n_q}^{q}\left(\mathbf{r}_{ij}\right)+\sum_{q\neq q'}\mathcal{E}_q\mathcal{E}_{q'}\left(\hat{e}_q\cdot\hat{e}_{q'}\right)\times\right.$$
$$\left.\times\left(e^{i\left(\mathbf{k}_{q'}-\mathbf{k}_q\right)\cdot\mathbf{r}}\widehat{\mathcal{AA}}_{m,n_q}^{q,\,q',-}-Adj.\right)\right\}. \quad (17)$$

Analogous expressions for the commutators of $\hat{\mathcal{H}}_{id}^{AD}$ and $\hat{\mathcal{H}}_{dd'}^{DD}$ with the intensity operator $\hat{\mathcal{I}}$ can be written, by introducing, for example, the notations in each case $\widehat{\mathcal{AD}}_{m,n_q}^{q}\left(\mathbf{r}_{id}\right)$, $\widehat{\mathcal{AD}}_{m,n_q}^{q,\,q',-}\left(\mathbf{r}_{id}\right)$, $\widehat{\mathcal{AD}}_{m,n_q}^{q,\,q',+}\left(\mathbf{r}_{id}\right)$ and $\widehat{\mathcal{DD}}_{m,n_q}^{q}\left(\mathbf{r}_{dd'}\right)$, $\widehat{\mathcal{DD}}_{m,n_q}^{q,\,q',-}\left(\mathbf{r}_{dd'}\right)$, $\widehat{\mathcal{DD}}_{m,n_q}^{q,\,q',+}\left(\mathbf{r}_{dd'}\right)$ in the place of $\widehat{\mathcal{AA}}_{m,n_q}^{q}\left(\mathbf{r}_{ij}\right)$, $\widehat{\mathcal{AA}}_{m,n_q}^{q,\,q',-}\left(\mathbf{r}_{ij}\right)$, $\widehat{\mathcal{AA}}_{m,n_q}^{q,\,q',+}\left(\mathbf{r}_{ij}\right)$, accordingly.

Using the commutation relations between the pairs of bosonic operators, we find

$$\left[\hat{\mathcal{H}}^{F},\hat{\mathcal{I}}\right]=$$
$$=\sum_{q,q'}\hbar\omega_q\left(\hat{e}_q\cdot\hat{e}_{q'}\right)\mathcal{E}_q\mathcal{E}_{q'}a_q^{\dagger}a_{q'}\left[e^{i\left(\mathbf{k}_{q'}-\mathbf{k}_q\right)\cdot\mathbf{r}}-\text{C.C.}\right]. \quad (18)$$

Using the above-found commutators (12)–(18), the parentheses of the commutation operation can be removed in the terms $\left[\hat{\mathcal{H}},\left[\hat{\mathcal{H}},\hat{\mathcal{I}}\right]\right]$ in (11), so that

$$\left[\hat{\mathcal{H}}'^{AF},\left[\hat{\mathcal{H}}'^{AF},\hat{\mathcal{I}}\right]\right]=$$
$$=-\sum_{\varsigma,D'}\sum_{i_\varsigma=1}^{N-2D'}\sum_{\ell,D}\sum_{i_\ell=1}^{N-2D}\sum_{q,\tilde{q}}\left(\mathcal{F}\hat{\mathbf{S}}_{i_\varsigma}\cdot\hat{e}_{\tilde{q}}\right)\left(\mathcal{F}\hat{\mathbf{S}}_{i_\ell}\cdot\hat{e}_q\right)\times$$
$$\times\left(\hat{e}_q\cdot\hat{e}_{\tilde{q}}\right)\mathcal{E}_q^2\mathcal{E}_{\tilde{q}}^2\left\{e^{i(\mathbf{k}_{\tilde{q}}-\mathbf{k}_q)\cdot\mathbf{r}}e^{i\mathbf{k}_q\cdot\mathbf{r}_{i_\ell}}e^{-i\mathbf{k}_{\tilde{q}}\cdot\mathbf{r}_{i_\varsigma}}+\text{C.C.}\right\}; \quad (19)$$

$$\left[\hat{\mathcal{H}}^{F},\left[\hat{\mathcal{H}}'^{AF},\hat{\mathcal{I}}\right]\right]=$$
$$=-\sum_{\ell,D}\sum_{i_\ell=1}^{N-2D}\sum_{q,\tilde{q}}\hbar\omega_{\tilde{q}}\left(\mathcal{F}\hat{\mathbf{S}}_{i_\ell}\cdot\hat{e}_q\right)\left(\hat{e}_q\cdot\hat{e}_{\tilde{q}}\right)\mathcal{E}_q^2\mathcal{E}_{\tilde{q}}\times$$
$$\times\left\{e^{i(\mathbf{k}_{\tilde{q}}-\mathbf{k}_q)\cdot\mathbf{r}}e^{i\mathbf{k}_q\cdot\mathbf{r}_{i_\ell}}a_{\tilde{q}}+Adj.\right\}. \quad (20)$$

We also expand the commutator of the atomic operator $\hat{\mathcal{H}}'^{A}$ with the found commutation relation (12), mainly

$$\left[\hat{\mathcal{H}}'^{A},\left[\hat{\mathcal{H}}'^{AF},\hat{\mathcal{I}}\right]\right]=$$
$$=\sum_{\ell,D}\sum_{i_\ell=1}^{N-2D}\left\{\mathcal{F}\hat{\mathbf{S}}_{i_\ell}\cdot\sum_{q,\tilde{q}}\hat{e}_q\mathcal{E}_q^2\left(\hat{e}_q\cdot\hat{e}_{\tilde{q}}\right)\mathcal{E}_{\tilde{q}}\times\right.$$
$$\times\left(e^{i(\mathbf{k}_{\tilde{q}}-\mathbf{k}_q)\cdot\mathbf{r}}e^{i\mathbf{k}_q\cdot\mathbf{r}_{i_\ell}}F_+a_{\tilde{q}}-e^{-i(\mathbf{k}_{\tilde{q}}-\mathbf{k}_q)\cdot\mathbf{r}}e^{-i\mathbf{k}_q\cdot\mathbf{r}_{i_\ell}}F_-a_{\tilde{q}}^{\dagger}\right)+$$
$$+\mathcal{F}\hat{\mathbf{S}}_{i_\ell}\cdot\sum_{q,\tilde{q}}\hat{e}_q\mathcal{E}_q^2\left(\hat{e}_q\cdot\hat{e}_{\tilde{q}}\right)\mathcal{E}_{\tilde{q}}\hbar\omega_q\times$$
$$\left.\times\left(e^{i(\mathbf{k}_{\tilde{q}}-\mathbf{k}_q)\cdot\mathbf{r}}e^{i\mathbf{k}_q\cdot\mathbf{r}_{i_\ell}}a_{\tilde{q}}+Adj.\right)\right\}. \quad (21)$$

Here, $F_{\pm}=\left[\omega_q^2\left(\frac{1}{c^2}\pm\frac{M_{i_\ell}}{\hbar\omega_0}\right)\frac{\hbar^2}{2M_{i_\ell}}\mp\frac{1}{2}\hbar\omega_q\right]$. We used the following relations and identities: $|\rangle_i\langle|_i\hat{\mathcal{H}}_i'^{A}|\rangle_i\langle|_i=$ $=-|\mathbf{r}_i\rangle\langle\mathbf{r}_i|\frac{\hbar^2\nabla_i^2}{2M_i}|\mathbf{r}_i\rangle\langle\mathbf{r}_i|+\hbar\omega_{\alpha_i}\sigma_i^{\dagger}\sigma_i$, $\left[\sigma_i^{\dagger}\sigma_i,\sigma_i\right]=-\sigma_i$, $\left[\sigma_i^{\dagger}\sigma_i,\sigma_i^{\dagger}\right]=\sigma_i^{\dagger}$, $\left[\sigma_i\sigma_i^{\dagger},\sigma_i\right]=\sigma_i$, and $\left[\sigma_i\sigma_i^{\dagger},\sigma_i^{\dagger}\right]=$ $=-\sigma_i^{\dagger}$. Here, $\sigma_i=|\alpha\rangle\langle\alpha'|_i$ with $\alpha,\,\alpha'\in\{a,b\}$. And,

$\left[\nabla_i^2, e^{i\mathbf{k}_q\cdot\mathbf{r}_i}\right] = e^{i\mathbf{k}_q\cdot\mathbf{r}_i}\left(-k^2 + 2i\mathbf{k}_q\cdot\nabla_i\right)$ was substituted into (21) with regard for (6).

The series of commutators in (11) can be continued in the fashion provided above.

Therefore, to compute the value of the absorption coefficient defined by (11), the distribution $\hat{\rho}_\Psi$ over the initial states $|\Psi\rangle_0$ of the system has to be given. Because of the quite complicated expressions for the commutators, the application of certain statistical properties of the system is not trivial. The quantitative example of the averaging procedure and the corresponding evaluation of the absorption coefficient will be discussed in the separate work.

4. Conclusion

The derived expression for the absorption coefficient is therefore based on the commutation relation between the operators of electric field and intensity. It is of interest to note that the contribution of (19) forms a non-linear dependence of the absorption coefficient on the atomic density even in the case of negligible binary interaction defined by $\hat{\mathcal{H}}^{AA}$. Under some conditions, this can be referred to a certain kind of collective effect. Thus, the quantization of the electromagnetic field, for example, in the optical region can play a fundamental role in defining the absorption/emission properties of a matter.

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УЗАГАЛЬНЕННЯ КОЕФІЦІЄНТА ПОГЛИНАННЯ ДЛЯ ГАЗУ АТОМІВ У ЗВ'ЯЗАНИХ СТАНАХ

Р е з ю м е

У даній роботі наведено приклад побудови коефіцієнта поглинання в термінах квантової оптики. Розроблена методика обчислення враховує квантування електромагнітного поля, просторові ступені вільності атомів, ефект Доплера, та парну взаємодію між частинками. Показано, що в основі квантово-оптичного формулювання коефіцієнта поглинання може лежати комутаційне співвідношення між операторами електричного поля та інтенсивності. Показано, що нелінійна залежність коефіцієнта поглинання від атомної концентрації, навіть у випадку нехтування бінарною взаємодією, може бути пояснена цілком визначеним типом квантово-оптичних колективних ефектів.