The structure of a solution of the generalized Maxwell–Bloch system of equations describing the strongly pumped interacting two-level atoms is discussed. This structure is represented by means of the corresponding differential equations for each contributing process. The interaction between the processes is introduced through the interaction integral and is illustrated by the specific system of graphs. The method allows one to describe the quantum-field-induced long-range interaction prevailing over short-range collisions and causing the broadening, narrowing, and shifts of an absorption line shape. The description is given in terms of the interaction integrals which couple the collective atomic polarization and population inversion. The contributions from different effects are analyzed with the use of the additivity of the corresponding absorption/reemission rates.

Keywords: two-laser beam spectroscopy, absorption coefficient, quantum optics, many-body interaction, non-linear optics.

1. Introduction

In this work, we investigate the analytical method of description of a many-atom system interacting with a strong pumping and counterpropagating scanning probe laser beams. The system is described by means of the corresponding system of non-linear integro-differential equations.

The problem of a complete consistent analytical description of the interacting many-body system is actual for developing the two-beam spectroscopy (so-called optical-optical double resonance, see, e.g., [1–3]). Mainly, the establishment of the adequate interpretation method is emphasized here. Therefore, some structural features of the generalized Maxwell–Bloch system of equations are revealed.

The model system under discussion takes the quantization of an electromagnetic field into account. In comparison with the semiclassical description (e.g., [4–17]), the quantum optics can uncover quantitatively and qualitatively new effects (see examples in [18–26]).

The well-known Maxwell–Bloch equations describe, strictly speaking, the model of interaction of the radiation field with a dilute ensemble of atoms in the semiclassical dipole approximation. The generalized Maxwell–Bloch system of equations for an optically dense medium is derived by the authors in [21]. The construction of the system of equations is based on the model Hamiltonian in the short time scale limit (see details in [27]). In comparison with other works (e.g., [28]), the relatively fast cooperative radiative atom-atom interactions are taken into account.

1 The paper was presented at the XXIVth Galyna Puchkowska International School-Seminar “Spectroscopy of Molecules and Crystals” (August 25–30, 2019, Odesa, Ukraine).
2. Model

2.1. Generalized Rabi frequency

Suppose that the ensemble of \( N \) atoms that fill a volume \( V \) optically transparent in the defined direction interacts with the modes of a quantized electromagnetic field of two oppositely directed lasers. The schematic illustration of the model gas of atoms (molecules) in a transparent chamber at the temperature \( T \) is illustrated in Fig. 1. The system is open (without mirrors). The two counterpropagating laser beams, strong and relatively weak, are passing through the sample. The frequencies of the beams are close to the optical resonant transition of the gas components. The emission spectrum of the model gas is narrow (within the double Rabi frequency of the strong beam), so that only two energy levels in the possible induced optical transitions are involved.

The frequencies of the modes of two electromagnetic fields are distributed near the resonant frequency \( \omega_0 \) of a single atomic transition between only two levels \( b \) (ground state) and \( a \) (excited state). The natural decay rate of an isolated atom from its excited state is defined by the quantity \( \gamma \). The interatomic and atom-field interactions are defined by the model Hamiltonian including the operators of the intrinsic energy of free atoms, energy of a free external field, and energy of the quantum optical dipole-dipole coupling between all quite close pairs of particles (see details in [27]). Based on the model Hamiltonian, the microscopic evolution equations can be built for the \( N \)-particle density matrix elements. As was shown in [21], the corresponding macroscopic kinetic equations in terms of the one-particle probability density matrix and the effective collective field can have the form of the generalized Maxwell–Bloch equations. Let \( \rho_{ba}(t,r) \) and \( \rho_{ab}(t,r) \) be the macroscopic probability densities to find an atom during the infinitesimal time interval \( (t-dt, t+dt) \) at the location of the physically small volume \( (r+dr) \) in the excited and ground states, accordingly; and let \( \rho_{ba}(t,r) = \rho_{ab}^{(0)}(t,r) \) be the polarization of the gas medium defining the averaged “off-diagonal” density matrix element for an atom. The appropriate modification of the common Bloch equations (see, e.g., [18], Ch. 5.4, pp. 165–167) is therefore taken into account in the following formula for a generalized Rabi frequency:

\[
\Omega(t,r) = \Omega^{AF}(t,r) + \Omega^{Int}(t,r),
\]

where \( \Omega^{AF}(t,r) = \frac{\hat{\varphi}\cdot E(t,r)}{\hbar} \). Here, the applied external field is approximated by the strong and weak counterpropagating monochromatic plane electromagnetic waves with Rabi couplings \( \Omega_1 \gg \Omega_2 \), accordingly, and slightly different frequencies \( \omega \) (for a pump) and \( \nu \) (for a signal): \( \hat{E}(t,r) = Er e^{-i(\omega t-k_1 r)} + Er e^{-i(\nu t+k_2 r)} \), where \( k_2 \parallel k_1 \).

The amplitudes \( E_1 \) and \( E_2 \) can depend on the spatial coordinates \( r \) and time \( t \), according to the Maxwell equations with the polarization \( P(t,r) \) of the system per unit volume, which is determined by the averaged “off-diagonal” density matrix element: \( P(t,r) = 2N\hat{\varphi}\hat{\varphi}^* \Re \{\rho_{ba}(t,r)\} \); \( \hat{\varphi}\hat{\varphi}^* \) is the off-diagonal matrix element of the dipole moment of an atom for the transition \( b \rightarrow a \), with \( \hat{\varphi}^* \) being the unit vector.

The interaction strength (or the so-called Rabi frequency for the effective collective field) is

\[
\Omega^{Int}(t,r) = -\frac{2}{\hbar} \chi \int dr' \left\{ \Re \{\rho_{ba}(t,r')\} Q(r,r') \right\},
\]

where \( \chi \approx \frac{1}{4\pi\varepsilon_0} N\varphi^2 \) with the integral kernel corresponding to the long-range dipole-dipole coupling (originating from the results in [27])

\[
Q(r,r') = \frac{\hat{\varphi}\hat{\varphi}' - |r-r'|^2}{|r-r'|^3} - 3 \left( \hat{\varphi} (r-r') \hat{\varphi}' (r-r') \right) |r-r'|^3.
\]

Note that, in accordance with the macroscopic definitions, the atomic concentration can be determined by the expression: \( n(r) = N (\rho_{aa}(t,r) + \rho_{bb}(t,r)) \).

3. Structure of the Solution

Suppose that the system evolves near a stationary state defined by the unperturbed diagonal and off-diagonal density matrix elements \( \rho^{(0)}_{\alpha\beta} \) for \( \alpha \in \{a, b\} \) and \( \beta \in \{a, b\} \).
In view of the weakness of the probe field and the interatomic coupling in comparison with the driving field, the inequalities

\[
\left| \Omega_2 \right| \ll 1 \quad \text{and} \quad \left| \Omega_{\text{int}} \right| \ll 1 \quad \text{with} \quad \Omega_2 = \frac{\varphi \Phi E_2}{\hbar} \quad (4)
\]

allow us to represent the solution of the macroscopic Maxwell–Bloch equations (see [21]) with the generalized Rabi frequency (1) through the following terms:

\[
\rho_{\alpha\beta}(t, r) = \rho_{\alpha\beta}^{(0)} + \delta \rho_{\alpha\beta}(t, r) + \delta \hat{\rho}_{\alpha\beta}(t, r) + + \delta \tilde{\rho}_{\alpha\beta}(t, r) + \delta \rho_{\alpha\beta}(t, r), \quad (5)
\]

where \( \rho_{\alpha\beta}^{(0)} \) with \( \alpha \in \{a, b\} \) and \( \beta \in \{a, b\} \) is the “equilibrium” solution for the system of pumped non-interacting atoms; \( \delta \rho_{\alpha\beta}(t, r) \) for \( \alpha \in \{a, b\} \), and \( \beta \in \{a, b\} \) is a perturbation to \( \rho_{\alpha\beta}^{(0)} \) in the case of non-zero interaction term \( \Omega_{\text{int}} \neq 0 \) and without external probe field \( \Omega_2 = 0 \). The correction \( \delta \tilde{\rho}_{\alpha\beta}(t, r) \) is induced, when both the strong and weak fields are applied without interatomic interaction \( \Omega_{\text{int}} = 0 \), and \( \delta \tilde{\rho}_{\alpha\beta}(t, r) \) takes into account the corrections to the absorption/emission rates induced by the perturbed population inversion and the polarization \( \delta \rho_{\alpha\beta}(t, r) \) (see [21] for details). Therefore, \( \Omega_2 \neq 0 \) and \( \Omega_{\text{int}} \neq 0 \), and \( \delta \tilde{\rho}_{\alpha\beta}(t, r) \) represents a strongly non-linear perturbation of the interaction term \( \Omega_{\text{int}} \delta \tilde{\rho}_{\alpha\beta} + \delta \tilde{\rho}_{\alpha\beta} + \delta \rho_{\alpha\beta} \) induced by the dipole-dipole coupling of the perturbed population and the polarization of the medium.

When the steady-state approximation is applied to the pumping field [in other words, when \( \rho_{\alpha\beta}^{(0)}, \rho_{\alpha\beta}^{(0)} \), and \( \rho_{\alpha\beta}^{(0)} \) are assumed to be essentially larger in their absolute values in comparison with the other terms in (5)], the integral Rabi frequency \( \Omega_{\text{int}} \) can be factorized to time and space functions. Namely,

\[
\Omega^\text{int}(t, r) \propto I(k, r), \quad (6)
\]

where

\[
I(k, r) = \int dr' e^{ik.(r-r')} Q(r, r'). \quad (7)
\]

Here, the integration is defined over the space volume \( V \) outside the spherical shell with the radius equal to the average distance between the nearest atoms.

The perturbations \( \delta \tilde{\rho}_{\alpha\beta}(t, r), \delta \tilde{\rho}_{\alpha\beta}(t, r) \) are defined as solutions of the corresponding systems of differential equations.

### 3.1. The case where the dipole “probe” field is much stronger than the dipole-dipole interaction

By definition, the matrix elements of \( \delta \tilde{\rho}(t, r) \) have to satisfy the following system of equations:

\[
\frac{\partial}{\partial t} \delta \tilde{\rho}_{aa}(t, r) = -\gamma \delta \tilde{\rho}_{aa}(t, r) + + \frac{i}{\hbar} \bigl[ \rho_{ba}^{(0)}(t, r) \tilde{\rho}_E(t) - c. c. \bigr] + + \frac{i}{\hbar} \bigl[ \tilde{\rho}_E(t) \tilde{\rho}_E^*(t) - (E_1(t) + E_2(t)) - c. c. \bigr]; \quad \delta \tilde{\rho}_{bb}(t, r) = -\delta \tilde{\rho}_{aa}(t, r); \quad (8)
\]

\[
\frac{\partial}{\partial t} \delta \tilde{\rho}_{ba}(t, r) = (-\gamma_{ba} + i \omega_0) \delta \tilde{\rho}_{ba}(t, r) + + \frac{i}{\hbar} \bigl[ \rho_{ba}^{(0)}(t, r) \tilde{\rho}_E(t) + \frac{i}{\hbar} \bigl[ \delta \tilde{\rho}_{aa}(t, r) - - \delta \tilde{\rho}_{bb}(t, r) \bigr] \tilde{\rho}_E(t) \tilde{\rho}_E^*(t) - (E_1(t) + E_2(t)) \bigr]; \quad (8)
\]

In this approximation, we do not account for the quasiclassical equation of motion for the averaged velocity of atoms. We recall that \( E_2(t, r) = E_{02} e^{-i(\nu t + k_2 r)} \).

### 3.2. The perturbation induced by the “interaction” of the “probe” field with the perturbation \( \delta \rho \) found without a probe field

The process (perturbation) \( \delta \tilde{\rho}(t, r) \) is defined as the solution of the system of equations as follows:

\[
\frac{\partial}{\partial t} \delta \tilde{\rho}_{aa}(t, r) = -\gamma \delta \tilde{\rho}_{aa}(t, r) + + \frac{i}{\hbar} \bigl[ \delta \rho_{aa}(t, r) \tilde{\rho}_E(t) - c. c. \bigr] + + \frac{i}{\hbar} \bigl[ \tilde{\rho}_E(t) \tilde{\rho}_E^*(t) - (E_1(t) + E_2(t)) - c. c. \bigr]; \quad (11)
\]

\[
\delta \tilde{\rho}_{bb}(t, r) = -\delta \tilde{\rho}_{aa}(t, r); \quad (12)
\]

\[
\frac{\partial}{\partial t} \delta \tilde{\rho}_{ba}(t, r) = (-\gamma_{ba} + i \omega_0) \delta \tilde{\rho}_{ba}(t, r) + + \frac{i}{\hbar} \bigl[ \delta \rho_{ba}(t, r) \tilde{\rho}_E(t) + \frac{i}{\hbar} \bigl[ \delta \tilde{\rho}_{aa}(t, r) - - \delta \tilde{\rho}_{bb}(t, r) \bigr] \tilde{\rho}_E(t) \tilde{\rho}_E^*(t) - (E_1(t) + E_2(t)) \bigr]. \quad (13)
\]
3.3. Corrections induced by the perturbation of the “interaction” integral

The perturbation (process) \( \delta \rho^{(2)} (t, r) \) is generated by the integral terms in the generalized Maxwell–Bloch system of equations (see details in [21]), when one considers the above-found perturbations under the integrals describing the quantum optical dipole-dipole contribution. Avoiding the resonant terms and keeping the introduced first-order perturbations under the “interaction” integral, we get that \( \delta \rho^{(2)} (t, r) \) is defined by the following system of equations:

\[
\frac{\partial}{\partial t} \delta \rho_{aa}^{(2)} (t, r) = -\gamma \delta \rho_{aa}^{(2)} (t, r) + \left( \frac{i}{\hbar} \delta \rho_{aa}^{(2)} (t, r) \right) \phi E_i (t, r) - c.c. \] + \frac{\chi}{\hbar} \left( \rho_0^0 e^{i(\omega t - k \cdot r)} \text{Int} (\delta \rho_{ba}^{(1)} ) + + \rho_{ba}^0 \left( \text{Int} (\delta \rho_{ba}^{(1)} ) \right)^* \right) + \rho_{ba} \delta \rho_{ba}^{(2)} (t, r) e^{i(\omega t - k \cdot r)} I + \left( \rho_{ba}^0 \delta \rho_{ba}^{(1)} (t, r) I + c.c. \right); \tag{14}
\]

where \( \delta \rho_{ba}^{(2)} (t, r) \) is defined by the integral Rabi frequency \(-\hbar^{-1} \text{Int} (\delta \rho_{ba}^{(1)} ) / 2\chi \), when the above-introduced perturbations are included (see more details in [21]).

3.3.1. Total absorption coefficient, probe transmittance and gain dependent on atomic density

Accordingly, if the time dependence of the probe amplitude is neglected in the case of the near-stationary system state (in the sense of dynamic equilibrium), then the appropriate Maxwell equation for the slowly varying probe field amplitude is as follows:

\[
\frac{\delta \rho_{aa}^{(2)} (t, r)}{\delta z} = \frac{1}{2} E_{02} (r) \alpha'_{tot}. \tag{17}
\]

where the total probe absorption coefficient \( \alpha'_{tot} \) is defined through the total absorption/re-emission rate denoted further as \( W'_{tot} \) at the given location.

3.4. Superposition of the induced absorption/reemission rates

Therefore, the total local absorption/reemission rate of probe photons, denoted as \( W'_{tot} \), is the sum of the rates induced by perturbations:

\[
W'_{tot} = W' + \delta \tilde{W}' + \delta W''_{tot}, \tag{18}
\]

where \( \delta W' = -2\Omega_2 \text{Im} (\tilde{C}') \), \( \delta \tilde{W}' = -2\Omega_2 \text{Im} (\tilde{C}'') \), \( \delta W''_{tot} = -2\Omega_2 \text{Im} (C'_{tot}) \). The polarization amplitudes \( \tilde{C}', \tilde{C}'', \) and \( C'_{tot} \) are defined by the off-diagonal \( (\alpha \neq \beta) \) elements of the perturbations \( \delta \rho_{aa} (t, r), \delta \rho_{ba} (t, r), \) and \( \delta \rho_{bb} (t, r) \), correspondingly, and are related to oscillations with the frequency of a probe laser \( \nu \).

All values of the coherence (polarization) amplitudes \( \tilde{C}', \tilde{C}'', \) and \( C'_{tot} \) are found by solving the corresponding system of algebraic equations, being a result of the substitution of the perturbations in the form of series of the induced modes (see details in [21]) into the corresponding systems of differential equations.

Then the total local coefficient \( \alpha'_{tot} \) for the probe field is defined as a superposition of the coefficients “induced” by perturbations:

\[
\alpha'_{tot} = KW'_{tot} = \delta \alpha' + \delta \alpha'' + \delta \alpha''_{tot}, \tag{19}
\]

where the correspondence between the constituent coefficients and the absorption/reemission rates is established by the same superscripts, \( K = \frac{\text{Doppl} f \text{Nh} \nu}{c(\text{Im}(E_{02}(r)))^2} \) with \( c \) and Doppl \( f \) denoting the speed of light in vacuum and the Doppler correction (partition), accordingly.

The above-provided systems of differential equations for the additive components of the absorption coefficient allow us to deduce the definite structure of the total solution. This is briefly discussed in the next section.

4. Schematic Representation of the System of Differential Equations

In accordance with the systems of differential equations introduced in the previous section, the structure of the solution is illustrated in the scheme in Fig. 2. Here, the dependence of a term (such as \( \delta \rho_{tot} \), \( \delta \rho_{tot}^{(2)} \)) on the other induced processes is shown by the dashed arrows joining the processes that generate the term. The type of interaction that involves the generating processes is shown to the left of the figure (left
near resonant optical absorption ordinate), while the types of the processes are cited at the right side of the figure (right ordinate). Each induced term is depicted by the horizontal vector. The circle with enclosed directed arrow depicts the interaction caused by the virtual photon exchange between the collective polarization/population induced by the laser beams with the wave-vectors $k_1$ and $k_2$. It is introduced by the interaction integral $I$. The semi-circle depicts the interaction of the type of dipole-field coupling. It is introduced by the scalar products between the vectors of the fields with the wave vectors $k_1$ and $k_2$ and the induced dipole moments of the collective polarizations. In principle, the directions and lengths of the vectors can be defined by their contributions (addition or subtraction). For this purpose, a scaling map for the corresponding absorption/re-emission rates has to be developed depending on the ordinates (processes and the type of interaction). The possibility to describe the laser beam scattering by a rearrangement of the terms as in Fig. 2 requires an additional investigation. For example, the introduced terms (processes) $\delta \rho_1$, ..., $\delta \rho^{(2)}$ are defined by the series of amplitudes that can compose a state vector in the certain basis (see details in [21]). However, the analytical definition of the processes in the form of the corresponding differential equations is more flexible and can cover a variety of effects. An example of a scattering process in terms of diagrams is given below.

The interesting fact is that the light beam with the wave vector $k_1$ or $k_2$ cannot pass through a sample under the given conditions, because of the quite high absorption rate by an isolated atom. The process of transmission can be due to a specific transparent (dressed) state of the medium. The latter, in a simplified case, is depicted in diagram 3. The ellipse with the enclosed directed spring depicts the non-linear interaction between two beams with the wave vectors $k_1$ and $k_2$ by the means of the induction of the population (polarization) collective waves (shown by the enclosed spring). The curved arrow depicts a laser beam with a wave-vector $k$.

In the applied theoretical approximations, the scaling network in the solution structure has to be in the accordance with the corresponding analytical solution. The example of the analytical solution of the system of equations (8)–(10) (see details of the approximation in [21]) along with the corresponding experimental observations for a sample of sodium vapor (see work [29]) is presented in graph 4. Figure 4 shows the “probe” gain of a vapor cell (the ratio of the transmitted and initial intensities) as a function of the probe detuning. The dashed line follows the analytically derived dependence with a resolution of 0.45 GHz. The pump is shifted by $-700$ MHz relative to the averaged resonant frequency of the $D_2$ sodium line. The effective beam path-length is $1/300$ m. The corresponding
number of atoms per cubic wavelength is 241 (at a temperature of about 617 K). The experimental data observed under the same conditions in work [29] are shown by the solid line. The maximum gain for the probe field occurs at a detuning by the approximately double Rabi frequency from the averaged resonant frequency.

We mention several different features of the theoretical and experimental line-shapes in Fig. 4. Despite the qualitatively good coincidence at the left and right wings and the main red shifted peak, the divergence of the curves at some locations is caused by the limitations of the applied approximations and the theoretically undefined factors related to the specificity of the experimental setup. Mainly, the dependence on the optical path length expressed by (17) is disregarded. Furthermore, the system of equations has been solved analytically using the linearization of the integral term for the term $\delta \rho$. Moreover, a more accurate theory should explicitly include the thermal motion of atoms that, in our opinion, can generate a much more complicated dependence on the resonant frequency because of the Doppler shift.

5. Conclusion

It is shown that, due to the quantization of the electromagnetic field, the structure of the solution for the corresponding generalized system of Maxwell–Bloch equations can be represented through the additive processes induced by the quantum optical dipole-field and dipole-dipole couplings. The structure of the solution is represented by means of the corresponding differential equations for each contributing process. The interaction between the processes is represented through the interaction integral and was illustrated by the system of directed graphs. In terms of the interaction integral coupling the collective atomic polarization and population inversion, the contribution from different effects can be analyzed using the additivity of the corresponding absorption/emission rates. The method allows us to describe the quantum-field-induced long-range interaction prevailing over short-range collisions, causing the broadening, narrowing, and shifts of a near-resonant absorption line shape.

If the experimental two-beam absorption line shape is given, the proposed representation can be used for the determination of the macroscopic collective interaction parameter $I$, i.e., the interaction integral. The example of an analytical solution corresponding to the schematic representation of the structure of the solution is provided. Within the discussed approximation, the theory can provide a quite good agreement with the experiment. Therefore, when the non-linear quantum optical collective coupling between atoms and the field defines the most prominent effect in the experiment, the proposed method can be used. At this point, the application of the structural representation requires further development and testing.


Received 03.10.19