Validity of Rate Equations for Zeeman Coherences for Analysis of Nonlinear Interaction of Atoms with Laser Radiation

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Abstract

In this paper we, to our knowledge, for the first time obtain the rate equations for Zeeman coherences in the broad line approximation and steady-state balance equations directly from optical Bloch equations without the use of the perturbation theory. The broad line approximation allows us to use the adiabatic elimination procedure in order to eliminate the optical coherences from the optical Bloch equations, but the steady-state condition allows us to derive the balance equations straightforward. We compare our approach with the perturbation theory approach as given previously and show that our approach is more flexible in analyzing various experiments. Meanwhile we also show the validity and limitations of the application of the rate equations in experiments with coherent atomic excitation, when either broad line approximation or steady-state conditions hold. Thus we have shown the basis for modeling the coherent atomic excitation experiments by using the relatively simple rate equations, provided that certain experimental conditions hold.
I. INTRODUCTION

Coherent effects in laser radiation interaction with atoms and molecules play a major role in the physics and chemistry. Applications, such as electromagnetically induced transparency [1], laser cooling [2, 3, 4], lasing without inversion [5], coherent population transfer [6], different nonlinear magneto optical effects [7], new methods for magnetometry [8, 9], coherent control of chemical reactions [10] and many other are widely used as a powerful research tools. Theoretical and experimental investigations of the coherent effects become increasingly important, as they open the way for more practical applications. Apart from some relatively simple cases, when direct solution of time dependant Schrodinger equation can be used [6], usually, when one speaks about the modelling of experiments with atomic coherent excitation, he means the so-called ”optical Bloch equations” (OBE), or Liouville equations [11, 12, 13] for quantum density matrix $\rho$. These involve both optical and Zeeman coherences created in an ensemble of atoms. Zeeman coherences are quite stable and therefore it is relatively easy to employ them practically – for example, Zeeman coherences are a basic ingredient of sub-Doppler and sub-recoil laser cooling mechanisms [2, 3, 4]. Optical coherences, on the other hand, are very sensitive to a variety of factors – collisions, finite laser line-width, laser light fluctuations – both in phase and in amplitude, and many other. This means that, in describing a wide variety of atomic excitation experiments one can neglect the optical coherences. It leads to the well known rate equations for Zeeman coherences [12, 14]. By saying ”rate equations” we mean that they do not couple Zeeman coherences to optical coherences.

Among the first ones to obtain the rate equations for Zeeman coherences by neglecting the optical coherences were C.Cohen-Tannoudji and J.P.Barrat in 1961 [14]. They used perturbation theory to obtain the rate equations in the so-called ”broad-line approximation” (BLA) [15]. These rate equations were obtained by considering the excitation with light from the spectral lamp and did not include the light induced transition effects into the analysis. They assumed intuitively that one can neglect the optical coherences in case of such an excitation. No mathematical arguments were provided, and the only justification of the used model was the good agreement between the theory and experiment. This lack of the rigorous mathematical argument was overcome later by C.Cohen-Tannoudji – with a slightly different approach, through the use of the perturbation theory and assuming the BLA [12]. The BLA means, that the spectral line-width of the laser light used in excitation of atomic transition $\Delta \omega$ is very large compared to the natural line-width $\Gamma$ of the atomic transition

$$\Delta \omega \gg \Gamma, \quad (1)$$

and the spacing between laser modes $\delta \omega$ is small compared to $\Gamma$

$$\delta \omega < \Gamma. \quad (2)$$

In this case different “Bennett holes”, burnt by the various modes in the Doppler profile, overlap, and the structure caused by different holes in the atomic response disappears. If, in addition, the modes cover all the velocity distribution, the atomic response does not depend on the velocity of translation motion of the atom and quantum density matrix $\rho$ refers to internal variables only. In order to use the perturbation theory, the following condition must be satisfied:

$$\Delta \omega \gg \Gamma, \Gamma_p, \quad (3)$$

2
where $\Gamma_p$ is connected to the time $T_p = \frac{1}{\Gamma_p}$ characterizing the evolution of density matrix $\rho$ under the effect of the coupling with the light beam. The rate equations for Zeeman coherences, obtained by considering the conditions (11) – (13), are often called the BLA equations.

In the past rate equations for Zeeman coherences in BLA were very successfully used to analyze numerous nonlinear magneto optical effects. These include, for example, the interaction of molecules with multimode laser radiation – nonlinear Hanle effect, quantum beats, beat resonance, alignment to orientation conversion in a magnetic field etc., see, for example [16, 17] and references therein. This approach from the viewpoint of implementation in the form of computer routines seems to be substantially less demanding technically than the OBE.

On the other hand, these rate equations for Zeeman coherences currently is not an often used approach to describe the laser radiation interaction with atomic gas. At first this seems obvious, as for the typical laser and atomic line-width the BLA conditions seem to be a very special case.

However recently we have applied these equations for description of some linear and nonlinear magneto-optical effects in stationary interaction of alkali atoms with a broad band diode laser radiation [18, 19, 20, 21]. In these cases the BLA clearly did not hold. Nevertheless, the agreement between simulation and experiment was good. The detailed analysis showed that for the use of BLA equations one does not always have to consider the BLA conditions – a rather striking result at a first sight. For example, in the case of a "steady-state" excitation there actually are no limitations for the use of the BLA equations except for the "steady-state" itself. The "steady-state" or stationary excitation means that the excitation light does not depend on time, which implies the same for the total density matrix $\rho(t)$ – and this is the case in the large number of coherent atomic excitation experiments.

What was the reason for such a good agreement between simulation and experiment in the above experiments with alkali atoms? After a detailed analysis it turned out, that the key factor was the fact, that the spectral line-width of radiation from the diode lasers was mainly determined by the phase fluctuations. The problem was, that the rigorous analysis of the limitations of the rate equations for Zeeman coherences in case of a noisy laser radiation seemed to be still lacking. On the other hand, there has been a large amount of work (see the overview in [22, 23]) dealing with the OBE when the exciting radiation has a finite line-width arising from the fluctuations – both in phase and in amplitude.

Thus in this paper we use the results obtained for the OBE and to our knowledge for the first time obtain the rate equations for Zeeman coherences directly from the OBE. We also compare our approach with the perturbation theory approach [12] and show the advantage of our approach. We analyze the limitations of usage of the rate equations for Zeeman coherences in conditions of noisy laser radiation – with an accent on analysis for nonlinear magneto-optical effects in atoms. In the limit of large angular momentum (molecular case) such an analysis, at least partially, was done previously in [24]. In this paper our goal is to fill in this gap for the atoms. The obtained results are in such a good agreement with experiment, that we feel, that the relatively simple rate equations’ approach (comparing to the conventional OBE approach) is far too often undeservedly neglected, when discussing the modelling of nonlinear magneto-optical effects in atoms.
II. BROAD BAND RADIATION INTERACTION WITH ATOMS.

A. Exciting light.

In our analysis of usage of rate equation for Zeeman coherences we will describe the exciting light classically by a fluctuating electric field $\mathbf{E}(t)$ polarized along the unit vector $\mathbf{e}$

$$\mathbf{E}(t) = \varepsilon(t)\mathbf{e} + \varepsilon^*(t)\mathbf{e}^*, \tag{4}$$

$$\varepsilon(t) = |\varepsilon_\omega| e^{-i\Phi(t)-i(\overline{\omega}-k_\omega v)t}. \tag{5}$$

We account for a shift $\overline{\omega} - k_\omega v$ in the laser frequency due to the Doppler effect – $v$ is the velocity of translation motion of atoms and $k_\omega$ is the wave vector of the exciting light. $\overline{\omega}$ is the center frequency of the spectrum, $|\varepsilon_\omega|$ is an amplitude of laser light field and $\Phi(t)$ is the fluctuating phase, which gives the spectrum of the radiation a finite bandwidth $\Delta \omega$. If the phase fluctuations are completely random, then the line-shape of the exciting light is Lorentzian. In the case of a laser this light corresponds to the single mode laser with randomly fluctuating phase. In the case of a spectral lamp this light corresponds to the lamp, where the dominant mechanism for the line-width broadening is determined by the collisions between the radiating atoms or molecules. Note, that for the single-mode laser the BLA condition (2) is not fulfilled.

The Rabi frequency $\Omega_R$ is determined by

$$\Omega_R = \frac{d \cdot |\varepsilon_\omega|}{\hbar}, \tag{6}$$

where $d$ is assumed to be the strongest atomic electric dipole moment for the transition (transitions) under consideration.

B. Optical Bloch equations.

We consider the dipole interaction of an atom with a laser field in presence of an external static magnetic field $\mathbf{B}$. We assume that the atomic center of mass moves classically, which means, that the only effect of the dipole interaction of the atom with a laser field is an excitation of a classically moving atom at the internal transitions. In this case the internal atomic dynamics is described by the semiclassical atomic density matrix $\rho$, which parametrically depends on the classical coordinates of the atomic center of mass. We consider atoms with definite velocity $\mathbf{v}$, illuminated by the exciting light (4), (5), resonant with the $g \leftrightarrow e$ transition, in presence of an external static magnetic field $\mathbf{B}$, which removes the degeneracy of the levels $g$ and $e$, so that now we consider Zeeman sublevels $g_i$ and $e_i$. In writing OBE, see for example (22)

$$i\hbar \frac{\partial \rho}{\partial t} = \left[ \tilde{H}, \rho \right] + i\hbar \dot{\rho}, \tag{7}$$

we consider only the relaxation $\dot{\rho}$ due to spontaneous emission. This means that we neglect other relaxation mechanisms, such as collisions, fly-through relaxation etc. This assumption means, that different velocity groups do not interact – the density of atoms is sufficiently low. For simplicity we also assume that the atomic transition forms a closed system – cycling
transition. In this case the spontaneous relaxation terms for a closed system for the density matrix elements $\rho_{g_i g_j}$, $\rho_{g_i e_j}$, $\rho_{e_i g_j}$, $\rho_{e_i e_j}$ are:

$$
\hat{R}\rho_{g_i g_j} = \sum_{e_i e_j} \Gamma_{g_i g_j}^{e_i e_j} \rho_{e_i e_j},
$$

$$
\hat{R}\rho_{g_i e_j} = -\frac{\Gamma}{2} \rho_{g_i e_j},
$$

$$
\hat{R}\rho_{e_i g_j} = -\frac{\Gamma}{2} \rho_{e_i g_j},
$$

$$
\hat{R}\rho_{e_i e_j} = -\Gamma \rho_{e_i e_j},
$$

where $\Gamma_{g_i g_j}^{e_i e_j}$ describes the spontaneous relaxation from $\rho_{e_i e_j}$ to $\rho_{g_i g_j}$ and $\Gamma$ describes the spontaneous relaxation from $e \rightarrow g$. For the closed system it is obvious that $\sum_{g_i g_j} \Gamma_{g_i g_j}^{e_i e_j} = \Gamma$.

Hamiltonian $\hat{H} = \hat{H}_0 + \hat{V}$ includes the unperturbed atomic Hamiltonian $\hat{H}_0$, which depends on the internal atomic coordinates: $\hat{H}_0 |\Psi_n\rangle = E_n |\Psi_n\rangle$, and the dipole interaction operator $\hat{V} = -\hat{d} \cdot \mathbf{E}(t)$, where $\hat{d}$ is the electric dipole operator. Writing OBE explicitly for the density matrix element $\rho_{ij}$, we get:

$$
\frac{\partial \rho_{ij}}{\partial t} = -\frac{i}{\hbar} \left[ \hat{H}_0, \rho_{ij} \right] + \frac{i}{\hbar} \left[ \hat{d} \cdot \mathbf{E}(t), \rho_{ij} \right] + \hat{R}\rho_{ij} =
$$

$$
= -i\omega_{ij} \rho_{ij} + \hat{R}\rho_{ij} +
$$

$$
+ \frac{i}{\hbar} \sum_k \langle i | \hat{d} \cdot \mathbf{e} | k \rangle \rho_{kj} + \frac{i}{\hbar} \varepsilon_k \sum_k \langle i | \hat{d} \cdot \mathbf{e}^* | k \rangle \rho_{kj} -
$$

$$
- \frac{i}{\hbar} \varepsilon_k \sum_k \langle k | \hat{d} \cdot \mathbf{e} | j \rangle \rho_{ik} - \frac{i}{\hbar} \varepsilon_k \sum_k \langle k | \hat{d} \cdot \mathbf{e}^* | j \rangle \rho_{ik},
$$

where $\omega_{ij} = \frac{E_i - E_j}{\hbar}$ denotes the Zeeman splitting of the levels $i$ and $j$. By choosing quantization axis to be parallel to the external static magnetic field $B$, all the dependence of the density matrix on the $B$ field is included in the splitting term $\omega_{ij}$. Thus we arrive to the following equations for the density matrix elements $\rho_{g_i g_j}$, $\rho_{g_i e_j}$, $\rho_{e_i g_j}$, $\rho_{e_i e_j}$:

$$
\frac{\partial \rho_{g_i g_j}}{\partial t} = \frac{i}{\hbar} \sum_{e_k} \langle g_i | \hat{d} \cdot \mathbf{e} | e_k \rangle \rho_{e_k g_j} + \frac{i}{\hbar} \varepsilon_k \sum_{e_k} \langle g_i | \hat{d} \cdot \mathbf{e}^* | e_k \rangle \rho_{e_k g_j} -
$$

$$
- \frac{i}{\hbar} \varepsilon_k \sum_{e_k} \langle e_k | \hat{d} \cdot \mathbf{e} | g_j \rangle \rho_{g_i e_k} - \frac{i}{\hbar} \varepsilon_k \sum_{e_k} \langle e_k | \hat{d} \cdot \mathbf{e}^* | g_j \rangle \rho_{g_i e_k} - i\omega_{gigj} \rho_{g_i g_j} + \sum_{e_i e_j} \Gamma_{g_i g_j}^{e_i e_j} \rho_{e_i e_j},
$$

$$
(10)
$$

$$
\frac{\partial \rho_{g_i e_j}}{\partial t} = \frac{i}{\hbar} \sum_{e_k} \langle g_i | \hat{d} \cdot \mathbf{e} | e_k \rangle \rho_{e_k e_j} + \frac{i}{\hbar} \varepsilon_k \sum_{e_k} \langle g_i | \hat{d} \cdot \mathbf{e}^* | e_k \rangle \rho_{e_k e_j} -
$$

$$
- \frac{i}{\hbar} \varepsilon_k \sum_{g_k} \langle g_k | \hat{d} \cdot \mathbf{e} | e_j \rangle \rho_{g_i g_k} - \frac{i}{\hbar} \varepsilon_k \sum_{g_k} \langle g_k | \hat{d} \cdot \mathbf{e}^* | e_j \rangle \rho_{g_i g_k} - i\omega_{giej} \rho_{g_i e_j} - \frac{\Gamma}{2} \rho_{g_i e_j},
$$

$$
(11)
$$
\[
\frac{\partial \rho_{g,g_j}}{\partial t} = \frac{i}{\hbar} \varepsilon \sum_{g_k} \langle e_i | d \cdot e | g_k \rangle \rho_{g_k,g_j} + \frac{i}{\hbar} \varepsilon^* \sum_{g_k} \langle e_i | d \cdot e^* | g_k \rangle \rho_{g_k,g_j} - \\
- \frac{i}{\hbar} \varepsilon \sum_{e_k} \langle e_k | d \cdot e | g_j \rangle \rho_{e_je_k} - \frac{i}{\hbar} \varepsilon^* \sum_{e_k} \langle e_k | d \cdot e^* | g_j \rangle \rho_{e_je_k} - i\omega_{g_je_k} \rho_{g_je_k} - \frac{\Gamma}{2} \rho_{g_je_k},
\]

(12)

\[
\frac{\partial \rho_{g,e_j}}{\partial t} = \frac{i}{\hbar} \varepsilon \sum_{g_k} \langle e_i | d \cdot e | g_k \rangle \rho_{g_k,e_j} + \frac{i}{\hbar} \varepsilon^* \sum_{g_k} \langle e_i | d \cdot e^* | g_k \rangle \rho_{g_k,e_j} - \\
- \frac{i}{\hbar} \varepsilon \sum_{g_k} \langle g_k | d \cdot e | e_j \rangle \rho_{g_je_j} - \frac{i}{\hbar} \varepsilon^* \sum_{g_k} \langle g_k | d \cdot e^* | e_j \rangle \rho_{g_je_j} - i\omega_{g_je_j} \rho_{g_je_j} - \Gamma \rho_{g_je_j},
\]

(13)

The matrix elements of the type \( \langle e_i | d \cdot e | g_j \rangle \) can be calculated using the standard angular momentum algebra [17, 25, 26].

Now, in order to eliminate the fast oscillations with optical frequency \( \bar{\omega} \), we make the following substitutions:

\[
\rho_{g,g_j} = \overline{\rho_{g,g_j}} = \rho_{g,g_j},
\]

\[
\rho_{g,e_j} = \overline{\rho_{g,e_j}} e^{i(\bar{\omega} - k \cdot \bar{v}) t + i\Phi(t)},
\]

\[
\rho_{e_i,g_j} = \overline{\rho_{e_i,g_j}} e^{-i(\bar{\omega} - k \cdot \bar{v}) t - i\Phi(t)},
\]

\[
\rho_{e_i,e_j} = \overline{\rho_{e_i,e_j}} = \rho_{e_i,e_j}.
\]

(14)

By using the rotating wave approximation [11] and neglecting terms with double optical frequency, we arrive to:

\[
\frac{\partial \overline{\rho_{g,g_j}}}{\partial t} = \frac{i}{\hbar} |\varepsilon| \sum_{e_k} \langle g_i | d \cdot e^* | e_k \rangle \overline{\rho_{g_je_k}} - \frac{i}{\hbar} |\varepsilon| \sum_{e_k} \langle e_k | d \cdot e | g_j \rangle \overline{\rho_{g_je_k}} - \\
- i\omega_{g_je_k} \rho_{g_je_k} + \sum_{g_je_j} \Gamma_{g_je_j} \rho_{g_je_j} - \frac{\Gamma}{2} \rho_{g_je_j},
\]

(15)

\[
\frac{\partial \overline{\rho_{g,e_j}}}{\partial t} = \frac{i}{\hbar} |\varepsilon| \sum_{e_k} \langle g_i | d \cdot e^* | e_k \rangle \overline{\rho_{ke,e_j}} - \frac{i}{\hbar} |\varepsilon| \sum_{g_k} \langle g_k | d \cdot e^* | e_j \rangle \overline{\rho_{gke_k}} - \\
- i(\bar{\omega} - k \cdot \bar{v} + \omega_{g_i,e_j}) \overline{\rho_{g_i,e_j}} - \frac{\Gamma}{2} \overline{\rho_{g_i,e_j}} - i \frac{\partial\Phi(t)}{\partial t} \overline{\rho_{g_i,e_j}},
\]

(16)

\[
\frac{\partial \overline{\rho_{e_i,g_k}}}{\partial t} = \frac{i}{\hbar} |\varepsilon| \sum_{g_k} \langle e_i | d \cdot e | g_k \rangle \overline{\rho_{g_je_j}} - \frac{i}{\hbar} |\varepsilon| \sum_{e_k} \langle e_k | d \cdot e | g_j \rangle \overline{\rho_{e_je_j}} + \\
+ i(\bar{\omega} - k \cdot \bar{v} - \omega_{e_i,g_k}) \overline{\rho_{e_ig_k}} - \frac{\Gamma}{2} \overline{\rho_{e_ig_k}} + i \frac{\partial\Phi(t)}{\partial t} \overline{\rho_{e_ig_k}},
\]

(17)

\[
\frac{\partial \overline{\rho_{e_i,e_j}}}{\partial t} = \frac{i}{\hbar} |\varepsilon| \sum_{g_k} \langle e_i | d \cdot e | g_k \rangle \overline{\rho_{g_je_j}} - \frac{i}{\hbar} |\varepsilon| \sum_{g_k} \langle g_k | d \cdot e^* | e_j \rangle \overline{\rho_{g_je_j}} - \\
- i\omega_{e_i,e_j} \rho_{e_i,e_j} - \Gamma \rho_{e_i,e_j},
\]

(18)
C. Atoms in a fluctuating optical field.

The equations (13) – (18) are stochastic differential equations with stochastic variable \( \frac{\partial \Phi(t)}{\partial t} \). In an experiment, as a rule, we deal with quantities that are averaged over the time intervals that are large in comparison with phase fluctuation time in the excitation light source, therefore we need to perform the statistical averaging of the above equations. In order to do that, we solve the equations (16) and (17) (with initial condition \( \rho_{g_i,j}(t_0) = \rho_{a_i,j}(t_0) = 0 \)) and then take a formal statistical average over the fluctuating phases:

\[
\frac{\partial \langle \rho_{g_i,j} \rangle}{\partial t} = \frac{i}{\hbar} \sum_{e_k} \langle g_i | d \cdot e^* | e_k \rangle \langle \rho_{e_k,j} \rangle - \frac{i}{\hbar} \sum_{e_k} \langle e_k | d \cdot e | g_j \rangle \langle \rho_{g_i,e_k} \rangle - i\omega_{g_i,j} \langle \rho_{g_i,j} \rangle + \sum_{e_i e_j} \Gamma_{g_i,j} \langle \rho_{e_i,e_j} \rangle, 
\]

(19)

\[
\langle \rho_{g_i,j} \rangle = \frac{i}{\hbar} \sum_{e_k} \langle g_i | d \cdot e^* | e_k \rangle \int_{t_0}^{t} e^{-i(\omega_{g_i,j} - \omega_{e_k,j})(t-t')} \langle \rho_{e_k,j}(t')e^{-i[\Phi(t)-\Phi(t')]} \rangle dt' - \frac{i}{\hbar} \sum_{g_k} \langle g_k | d \cdot e^* | e_j \rangle \int_{t_0}^{t} e^{-i(\omega_{g_k,j} - \omega_{e_i,j})(t-t')} \langle \rho_{g_k,j}(t')e^{-i[\Phi(t)-\Phi(t')]} \rangle dt',
\]

(20)

\[
\langle \rho_{e_i,j} \rangle = \frac{i}{\hbar} \sum_{g_k} \langle e_i | d \cdot e | g_k \rangle \int_{t_0}^{t} e^{-i(\omega_{e_i,j} + \omega_{e_i,j})(t-t')} \langle \rho_{g_k,j}(t')e^{-i[\Phi(t)-\Phi(t')]} \rangle dt' - \frac{i}{\hbar} \sum_{e_k} \langle e_k | d \cdot e | g_j \rangle \int_{t_0}^{t} e^{-i(\omega_{e_k,j} + \omega_{e_i,j})(t-t')} \langle \rho_{e_k,j}(t')e^{-i[\Phi(t)-\Phi(t')]} \rangle dt',
\]

(21)

\[
\frac{\partial \langle \rho_{e_i,j} \rangle}{\partial t} = \frac{i}{\hbar} \sum_{g_k} \langle e_i | d \cdot e^* | g_k \rangle \langle \rho_{g_k,j} \rangle - \frac{i}{\hbar} \sum_{g_k} \langle g_k | d \cdot e^* | e_j \rangle \langle \rho_{e_k,g_k} \rangle - i\omega_{e_i,j} \langle \rho_{e_i,j} \rangle - \Gamma \langle \rho_{e_i,e_j} \rangle.
\]

(22)

Now we employ the relation (3) which allows us to use the decorrelation approximation (28, 29, 30). The decorrelation approximation means that we neglect the fluctuations of \( \rho_{a_i,j}(t) \) (\( a = e, g \)) around their mean value \( \langle \rho_{a_i,j}(t) \rangle \), and thus separate atom and field variables in (20) and (21):

\[
\langle \rho_{a_i,j}(t')e^{\pm i[\Phi(t)-\Phi(t')]} \rangle = \langle \rho_{a_i,j}(t') \rangle \langle e^{\pm i[\Phi(t)-\Phi(t')]} \rangle,
\]

(23)

where \( a = e, g \). The decorrelation approximation in general is valid only for Wiener-Levy-type (see below) phase fluctuations (30, 31). In the case of a general stochastic field the decorrelation approximation can be used as a first approximation only for weak fields below saturation (29, 30).
In order to evaluate the correlation function $\langle e^{\pm i[\Phi(t) - \Phi(t')]} \rangle$, we assume two simple models, which lead to similar results. The first one is the "phase jump" model \([32, 33, 34]\), which assumes the phase to remain constant, except sudden random "jumps", when it changes to a new constant value. This model is used to describe the fluctuations from the spectral lamp, when the dominant mechanism for the line-width broadening is determined by the collisions between the radiating atoms or molecules \([22, 32, 33]\). The second model is the "phase diffusion" model \([30, 31, 35, 36]\), which assumes the continuous random diffusion of the phase. This model is used to describe the fluctuations from the single-mode laser with fluctuating phase \([22, 35, 36]\).

1. "Phase jump" model.

Our analysis of phase jumps in excitation radiation is based on the detailed analysis of the model in case of optical Bloch equations performed in \([32, 33, 34]\). In this work we closely follow an approach, that is analogous to the effect of instantaneous collisions on the density matrix \([22]\). The random jump process is Poissonian in nature – the probability for the phase to change \(N\) times during time \(t - t'\) is

\[
P_N = \frac{1}{N!} \left( \frac{t - t'}{T} \right)^N e^{-\frac{t - t'}{T}},
\]

where \(T\) is the average time between successive phase jumps. Now we define $\langle e^{\pm i[\Phi(t) - \Phi(t')]} \rangle$ as the average phase change during one jump. If during the time \(t - t'\) there has been only one phase jump, then $\langle e^{\pm i[\Phi(t) - \Phi(t')]} \rangle_1 = \langle e^{\pm i\Delta \Phi} \rangle$. Obviously, if during the time \(t - t'\) there has been \(N\) phase jumps, then $\langle e^{\pm i[\Phi(t) - \Phi(t')]} \rangle_N = \langle e^{\pm i\Delta \Phi} \rangle^N$, as every jump in average adds one more multiplier $\langle e^{\pm i\Delta \Phi} \rangle$. In order to get the final expression for $\langle e^{\pm i[\Phi(t) - \Phi(t')]} \rangle$, we must average over every possible number \(N = 0 \div \infty\) of phase jumps during time \(t - t'\):

\[
\langle e^{\pm i[\Phi(t) - \Phi(t')]} \rangle = \sum_{N=0}^{\infty} P_N \langle e^{\pm i[\Phi(t) - \Phi(t')]} \rangle^N = e^{-\frac{t - t'}{T}} \sum_{N=0}^{\infty} \frac{1}{N!} \left( \frac{t - t'}{T} \langle e^{\pm i\Delta \Phi} \rangle \right)^N = e^{-\frac{t - t'}{T}} (1 - \langle e^{\pm i\Delta \Phi} \rangle).
\]

At this point we make further simplifications. We consider the case, when there is no correlation of the phase values before and after the jump \([32]\). Then \(T\) is also the correlation time of the phase (in average after the time \(T\) the phase "forgets" its past) \(T = \frac{2}{\Delta \omega}\). We also consider, that all the phase values occur with equal probability. Then $\langle e^{\pm i\Delta \Phi} \rangle = 0$, and \([25]\) becomes

\[
\langle e^{\pm i[\Phi(t) - \Phi(t')]} \rangle = e^{-\frac{\Delta \omega}{2} (t - t')}.
\]

This means that the spectral distribution of the exciting light is Lorentzian with FWHM $\Delta \omega$.

2. "Phase diffusion" model.

Our analysis of the influence of phase diffusion of the excitation radiation on the interaction of laser radiation with atoms we base on the phase diffusion model analyzed in
In phase diffusion model the field has a constant amplitude, but its phase is a fluctuating quantity, which obeys the Langevin equation

$$\frac{d\Phi(t)}{dt} = \varsigma(t), \quad (27)$$

where $\varsigma(t)$ is a Gaussian random force with correlation function

$$\langle \varsigma(t)\varsigma(t') \rangle = b\beta e^{-\beta|t-t'|}, \quad (28)$$

$$\langle \varsigma(t) \rangle = 0, \quad (29)$$

which means, that $\varsigma(t)$ obeys the Langevin equation for Brownian motion

$$\frac{d}{dt}\varsigma(t) + \beta\varsigma(t) = F(t), \quad (30)$$

where $F(t)$ is a $\delta$-correlated Gaussian force fulfilling the correlation

$$\langle F(t)F(t') \rangle = 2b\beta^2\delta(t-t'). \quad (31)$$

The meaning of the parameters $b$ and $\beta$ can be interpreted from the equations (27) – (31). $\frac{1}{\beta}$ is the correlation time of the phase time derivative $\varsigma(t)$, but $b$ gives the band-width of the field in the limit $\beta \to \infty$. Explicit expressions for $\beta$ and $b$ in terms of fundamental laser constants are discussed by Haken in [37], see also [38].

The spectrum of the exciting radiation, described by (27) – (31), is given by the Fourier transform of the correlation function

$$\langle e^{\pm i[\Phi(t) - \Phi(t')]\rangle} = \exp\left[-b|t-t'| + \frac{1}{\beta}(e^{-\beta|t-t'|} - 1)\right]. \quad (32)$$

For $\beta \gg b$ the spectrum is Lorentzian with FWHM $\Delta\omega = 2b$ and having a cut-off at frequencies $\beta$, but for $\beta \ll b$ the spectrum is Gaussian with FWHM $\sqrt{8\ln(2)b}\beta$.

In the limit $\beta \to \infty$ the spectrum is pure Lorentzian with FWHM $\Delta\omega = 2b$ and $\varsigma(t)$ becomes $\delta$-correlated

$$\langle \varsigma(t)\varsigma(t') \rangle = 2b\delta(t-t'), \quad (33)$$

but the phase $\Phi(t)$ obeys a Wiener-Levy stochastic process. As mentioned above, the Wiener-Levy process is the only one, for which the decorrelation approximation is mathematically rigorous [29, 31, 33]. It is easily understood, as in this process the relevant fluctuating quantity $\varsigma(t)$ is $\delta$-correlated (correlation time $\frac{1}{\beta}$ of $\varsigma(t)$ tends to zero when $\beta \to \infty$), and thus we can always separate the time-scales of evolution of $\langle \rho_{ai,aj}(t') \rangle$ and $\langle e^{\pm i[\Phi(t) - \Phi(t')]\rangle}$ in (23). Wiener-Levy stochastic process is a nonstationary Markov Gaussian process [27], and is described by the Langevin equation for Brownian motion with negligible acceleration [22, 27, 30, 31], which can be shown to be equivalent to the diffusion equation [22]. For Wiener-Levy process the relation (32) becomes:

$$\langle e^{\pm i[\Phi(t) - \Phi(t')]\rangle} = \exp[-b|t-t'|] = e^{-\frac{\Delta\omega}{2}(t-t')}, \quad (34)$$
where we have used the fact, that $t \gg t'$.

The Lorentz profile is not a good description of the wings of any laser spectrum, thus this model is appropriate for rather small detunings. However, as shown in [35, 36], for $\beta \gg b$ (or $\beta \gg \Delta \omega$) in the limit $\beta \gg \Gamma, \Omega_R$, the line-shape of the exciting light is Lorentzian with cut-off at frequencies $\beta$. In this case the damping term $\frac{\Delta \omega}{2}$ is simply multiplied by the cut-off term, dependent on the detuning [35, 36]. This corresponds to a more realistic model of the laser spectrum. We also have to remember, that for the rotating wave approximation $\omega_0 \gg \beta$, as $\beta$ is the correlation time of the phase time derivative $\zeta(t) = \frac{d\Phi(t)}{dt}$.

D. The effective relaxation caused by the fluctuations of the exciting light.

As can be seen, both approaches give similar results for time average phase fluctuation value - the effect of the phase fluctuations on the density matrix is simply to add the additional relaxation term, equal to the HWHM of the exciting light. Now we use (26) and (31) to rewrite (19) - (22) (for simplicity we further drop the averaging brackets):

$$\frac{\partial \rho_{g_i g_j}}{\partial t} = \frac{i}{\hbar} |\varepsilon_i| \sum_{e_k} \langle g_i | \mathbf{d} \cdot \mathbf{e}^* | e_k \rangle \tilde{\rho}_{e_k g_j} - \frac{i}{\hbar} |\varepsilon_i| \sum_{e_k} \langle e_k | \mathbf{d} \cdot \mathbf{e} | g_j \rangle \tilde{\rho}_{g_i e_k} - \frac{i \omega_{gigj}}{\hbar} \rho_{g_i g_j} + \sum_{e_i e_j} \Gamma_{e_i e_j} \rho_{e_i e_j},$$

(35)

$$\tilde{\rho}_{g_i e_j} = \frac{i}{\hbar} |\varepsilon_i| \sum_{e_k} \langle g_i | \mathbf{d} \cdot \mathbf{e}^* | e_k \rangle \int_{t_0}^{t} e^{-i \left(\frac{\omega_{e_i e_j}}{2} - \frac{\omega_{\Delta \omega}}{2}\right)(t-t')} \rho_{e_k e_j}(t') dt' - \frac{i}{\hbar} |\varepsilon_i| \sum_{g_k} \langle g_k | \mathbf{d} \cdot \mathbf{e}^* | e_j \rangle \int_{t_0}^{t} e^{-i \left(\frac{\omega_{g_k g_j}}{2} - \frac{\omega_{\Delta \omega}}{2}\right)(t-t')} \rho_{g_i g_k}(t') dt',$$

(36)

$$\tilde{\rho}_{e_i g_j} = \frac{i}{\hbar} |\varepsilon_i| \sum_{g_k} \langle e_i | \mathbf{d} \cdot \mathbf{e} | g_k \rangle \int_{t_0}^{t} e^{-i \left(\frac{\omega_{e_i e_j}}{2} - \frac{\omega_{\Delta \omega}}{2}\right)(t-t')} \rho_{g_k e_j}(t') dt' - \frac{i}{\hbar} |\varepsilon_i| \sum_{e_k} \langle e_k | \mathbf{d} \cdot \mathbf{e} | g_j \rangle \int_{t_0}^{t} e^{-i \left(\frac{\omega_{g_i g_j}}{2} - \frac{\omega_{\Delta \omega}}{2}\right)(t-t')} \rho_{e_i e_k}(t') dt',$$

(37)

$$\frac{\partial \rho_{e_i e_j}}{\partial t} = \frac{i}{\hbar} |\varepsilon_i| \sum_{g_k} \langle e_i | \mathbf{d} \cdot \mathbf{e} | g_k \rangle \tilde{\rho}_{g_k e_j} - \frac{i}{\hbar} |\varepsilon_i| \sum_{g_k} \langle g_k | \mathbf{d} \cdot \mathbf{e}^* | e_j \rangle \tilde{\rho}_{e_i g_k} - \frac{i \omega_{e_i e_j}}{\hbar} \rho_{e_i e_j} - \Gamma \rho_{e_i e_j}.$$  

(38)
III. RATE EQUATIONS.

For the sake of simplicity we further assume (with $\omega_B$ characterizing the Zeeman splitting):

$$\Delta \omega \gg \omega_B,$$

though this condition can be avoided at the expense of complication of the final rate equations. \[39\] means, that we can write:

$$\left( \frac{\Gamma}{2} + \frac{\Delta \omega}{2} \right) + i \left( \omega - k_\omega v + \omega_{giej} \right) \approx \left( \frac{\Gamma}{2} + \frac{\Delta \omega}{2} \right) + i \left( \omega - k_\omega v - \omega_0 \right),$$

(40)

$$\left( \frac{\Gamma}{2} + \frac{\Delta \omega}{2} \right) - i \left( \omega - k_\omega v - \omega_{eigj} \right) \approx \left( \frac{\Gamma}{2} + \frac{\Delta \omega}{2} \right) - i \left( \omega - k_\omega v - \omega_0 \right).$$

(41)

At this point we go further and assume certain conditions, which allows us to simplify significantly the expressions for optical coherences \[36\] and \[37\]. These conditions are either BLA \[11 - 3\] or the steady-state \((40)\) - see below) conditions. Under these circumstances the expressions for optical coherences \[36\] and \[37\] become:

$$\tilde{\rho}_{giej} = \frac{i}{\hbar \left( \frac{\Gamma}{2} + \frac{\Delta \omega}{2} \right) + i \left( \omega - k_\omega v - \omega_0 \right)} \left( \sum_{e_k} \langle g_i | d_1 \cdot e^* | e_k \rangle \langle e_k | d_1 \cdot e | g_j \rangle \rho_{e_k e_j} - \sum_{g_k} \langle g_k | d_1 \cdot e^* | e_j \rangle \rho_{g_k g_j} \right),$$

(42)

$$\tilde{\rho}_{eigj} = \frac{i}{\hbar \left( \frac{\Gamma}{2} + \frac{\Delta \omega}{2} \right) - i \left( \omega - k_\omega v - \omega_0 \right)} \left( \sum_{g_k} \langle e_j | d_1 \cdot e | g_k \rangle \rho_{g_k g_j} - \sum_{e_k} \langle e_k | d_1 \cdot e | g_j \rangle \rho_{e_k e_j} \right).$$

(43)

Now, by substituting \[42\] and \[43\] in \[36\] and \[38\] we arrive to the final rate equations:

$$\frac{\partial \rho_{g_k g_j}}{\partial t} = \Gamma_p \sum_{e_k, e_m} \langle g_i | d_1 \cdot e^* | e_k \rangle \langle e_m | d_1 \cdot e | g_j \rangle \rho_{e_k e_m} -$$

$$- \left( \frac{\Gamma_p}{2} + i \Delta E_p \right) \sum_{e_k, g_m} \langle g_i | d_1 \cdot e^* | e_k \rangle \langle e_k | d_1 \cdot e | g_m \rangle \rho_{g_m g_j} -$$

$$- \left( \frac{\Gamma_p}{2} - i \Delta E_p \right) \sum_{e_k, g_m} \langle e_k | d_1 \cdot e^* | e_k \rangle \langle g_m | d_1 \cdot e | g_j \rangle \rho_{g_k g_m} -$$

$$- i \omega_{giej} \rho_{g_k g_j} + \sum_{e_i} \rho_{e_i e_j} \Gamma_{e_i e_j} \rho_{e_i e_j},$$

(44)

$$\frac{\partial \rho_{e_k e_j}}{\partial t} = \Gamma_p \sum_{g_k, g_m} \langle e_i | d_1 \cdot e | g_k \rangle \langle g_m | d_1 \cdot e^* | e_j \rangle \rho_{g_k g_m} -$$

$$- \left( \frac{\Gamma_p}{2} - i \Delta E_p \right) \sum_{g_k, e_m} \langle e_i | d_1 \cdot e | g_k \rangle \langle g_k | d_1 \cdot e^* | e_m \rangle \rho_{e_m e_j} -$$

$$- \left( \frac{\Gamma_p}{2} + i \Delta E_p \right) \sum_{g_k, e_m} \langle e_m | d_1 \cdot e | g_k \rangle \langle g_k | d_1 \cdot e^* | e_j \rangle \rho_{e_i e_m} -$$

$$- i \omega_{e_i e_j} \rho_{e_i e_j} - \Gamma \rho_{e_i e_j},$$

(45)
where \( \mathbf{d}_1 = \frac{\mathbf{d}}{|\mathbf{d}|} \) denotes the electric dipole moment unity vector, and thus matrix elements \( \langle e_i | \mathbf{d} \cdot \mathbf{e} | g_j \rangle \) are written as:

\[
\langle e_i | \mathbf{d} \cdot \mathbf{e} | g_j \rangle = \langle e_i | \mathbf{d}_1 \cdot \mathbf{e} | g_j \rangle \langle e||d||g \rangle ,
\]

where \( \langle e||d||g \rangle \) is the so-called reduced dipole matrix element. Note, that for the steady-state situation we must consider condition (60) in the above equations. \( \Gamma_p \) and \( \Delta E_p \) are defined as:

\[
\begin{align*}
\frac{\Gamma_p}{2} &= \frac{|\varepsilon\omega|^2}{\hbar^2} \times |\langle e||d||g \rangle|^2 \times \frac{(\frac{\Gamma}{2} + \frac{\Delta \omega}{2})}{(\frac{\Gamma}{2} + \frac{\Delta \omega}{2})^2 + (\omega - k\sigma v - \omega_0)^2}, \\
\Delta E_p &= \frac{|\varepsilon\omega|^2}{\hbar^2} \times |\langle e||d||g \rangle|^2 \times \frac{\omega - k\sigma v - \omega_0}{(\frac{\Gamma}{2} + \frac{\Delta \omega}{2})^2 + (\omega - k\sigma v - \omega_0)^2}.
\end{align*}
\]

\( \Gamma_p \) is the probability per unit time of an absorption or stimulated emission process, and \( \Delta E_p \) describes the light shifts [14] produced by the light irradiation (dynamic Stark shift). For BLA conditions (1) – (3) (47) and (48) become:

\[
\begin{align*}
\frac{\Gamma_p}{2} &\approx \frac{|\varepsilon\omega|^2}{\hbar^2} \times |\langle e||d||g \rangle|^2 \times \frac{\Delta \omega}{(\Delta \omega)^2 + (\omega - k\sigma v - \omega_0)^2}, \\
\Delta E_p &\approx \frac{|\varepsilon\omega|^2}{\hbar^2} \times |\langle e||d||g \rangle|^2 \times \frac{\omega - k\sigma v - \omega_0}{(\Delta \omega)^2 + (\omega - k\sigma v - \omega_0)^2}.
\end{align*}
\]

Note also, that the phase fluctuations (described by the above models) reduce the saturation on resonance \((\omega - k\sigma v - \omega_0 = 0)\) by the factor \( \frac{\Gamma}{\Gamma + \Delta \omega} \), and increase the saturation far-off resonance \((\omega - k\sigma v - \omega_0 \gg \Gamma, \Delta \omega)\) by the factor \( \frac{\Gamma + \Delta \omega}{\Gamma} \).

When the density matrix for the excited state is calculated, one can obtain fluorescence intensity with specific polarization along the unit vector \( \mathbf{e}_1 \) as [14, 17, 39]:

\[
I(e_1) = \tilde{I}_0 \sum_{g_i,e_i,e_j} \langle e_i | \mathbf{d} \cdot \mathbf{e}_1^* | g_i \rangle \langle g_i | \mathbf{d} \cdot \mathbf{e}_j | e_j \rangle \rho_{e_i,e_j},
\]

where \( \tilde{I}_0 \) is a proportionality coefficient.

**IV. ANALYSIS AND CONCLUSIONS.**

**A. Perturbation theory approach.**

The obtained rate equations for Zeeman coherences coincide with equations obtained earlier in perturbation theory approach in [12]. In perturbation theory approach as small parameters are used ratios between rate constants involved in the problem \( (\Gamma_p, \Gamma) \) and line-width of the excitation radiation \( \Delta \omega \). Here we would like to stress that, however the obtained equations coincide, the approach used in this study is different and allows us to examine in more detail the limits of usage of rate equations for Zeeman coherences to analyze specific
experiments. To compare both approaches, let us have a brief look in method used and conclusions obtained with perturbation theory.

Let \( T_p = \frac{1}{\Gamma_p} \) be the time characterizing the evolution of density matrix \( \rho \) under the effect of the coupling with the light beam. In the following analysis it is assumed that the intensity is sufficiently low so that \( T_p \) is much longer than the correlation time \( T = \frac{1}{\Delta \omega} \) of the light wave

\[
\Delta \omega \gg \Gamma_p.
\] (52)

Now consider a time interval \( \Delta t \) such that

\[
T_p, \tau \gg \Delta t \gg T,
\] (53)

where \( \tau = \frac{1}{\Gamma} \). Since \( T_p, \tau \gg \Delta t \), one can conclude that \( \rho(t+\Delta t) - \rho(t) \) is very small and can be calculated by perturbation theory. By using perturbation theory, it is shown in [12], that the average variation of \( \rho \), \( \langle \rho(t+\Delta t) - \rho(t) \rangle \) (the average is taken over all possible values of the random function \( \varepsilon(t) \) - see below) is linear in \( \Delta t \) and only depends on \( \rho(t) \)

\[
\langle \rho(t+\Delta t) - \rho(t) \rangle = \frac{\Delta \rho(t)}{\Delta t}.
\] (54)

This means, that we can replace \( \frac{\Delta \rho(t)}{\Delta t} \) with the time derivative \( \frac{d\rho(t)}{dt} \), provided that we never use \( \frac{d\rho(t)}{dt} \) to describe the changes of \( \rho(t) \) over time intervals that are shorter than correlation time \( T \) of the light wave, which drives the atoms. \( \frac{\Delta \rho(t)}{\Delta t} = \frac{d\rho(t)}{dt} \) is called the “coarse grained” derivative [40].

In [12], the exciting light is taken to be the superposition of parallel plane waves having all the same polarization \( e \), but different amplitudes \( |\varepsilon_\mu| \), frequencies \( \omega_\mu \) and phases \( \Phi_\mu \)

\[
E(t) = \varepsilon(t)e + \varepsilon^*(t)e^*,
\] (55)

\[
\varepsilon(t) = \sum_\mu |\varepsilon_\mu| e^{-i\Phi_\mu - i(\omega_\mu - k\omega)v)t}.
\] (56)

BLA relations (1) – (3) hold and the relative phases of the different modes are assumed to be completely random and thus obeying the correlation relation:

\[
\langle e^{-i(\Phi_\mu - \Phi_\mu')} \rangle = \delta_{\mu\mu'},
\] (57)

The instantaneous electric field \( \varepsilon(t) \) of the light wave thus may be considered as a stationary random function, which obeys the correlation relation:

\[
\langle \varepsilon(t)\varepsilon^*(t-\tau) \rangle = \sum_{\mu,\mu'} |\varepsilon_\mu| |\varepsilon_{\mu'}| \langle e^{-i(\Phi_\mu - \Phi_{\mu'})} \rangle e^{-i(\omega_\mu - k\omega)v)t} e^{i(\omega_{\mu'} - k\omega)v)(t-\tau)} = \sum_{\mu} |\varepsilon_\mu|^2 e^{-i(\omega_\mu - k\omega)v)(t-\tau)}.
\] (58)

Applying perturbation theory, after some calculations with the consideration of (58) and “coarse grained” derivative, the rate equations are obtained [12], again considering condition
for simplicity. The obtained rate equations are exactly the same as the above derived equations (44), (45), but with $\Gamma_p$ and $\Delta E_p$ having slightly different form as defined in (47) – (50). This mismatch is easily avoided, if instead of the exciting light model (55) – (58) we take the model (1), (5), (26), (34). Then the rate equations are the same as equations (44), (45), with $\Gamma_p$ and $\Delta E_p$ defined as in (49) and (50).

Thus the perturbation theory approach is summarized as follows: we define "a priori" the BLA conditions (1) – (3) and then use the perturbation theory to obtain the rate equations - and thus we are restricted to the BLA case.

However, in our approach we obtain the "phase-averaged" OBE and then it is possible to choose between the BLA (1) – (3) or steady-state (60) possibilities.

Thus we arrive to the conclusion stated above that the approach discussed in this article allows to examine the limits of usage of rate equations for Zeeman coherences to a greater detail than the perturbation theory approach. Therefore it can be applied to analyze larger number of experimental situations.

B. Velocity dependence.

When we look at (47) – (50), we see, that $\Gamma_p$ (induced transition rate) and $\Delta E_p$ (dynamic Stark shift) are velocity dependent, and thus are also the equations (44), (45). This means, that in describing the observable signal we need to take into account all the velocity groups involved (note, that we have already assumed, that different velocity groups do not interact – the density of atoms is sufficiently low). In standard method one has to determine the signal dependence on velocity and then sum (integrate) over the velocities (of course, assuming that velocity distribution is known). However, usually the signal dependence on velocity cannot be found in analytical form, as can be seen from (44) – (50). Thus a large amount of calculation is necessary to determine this dependence – and still it is just an approximation.

The situation is simplified only for a specific kind of experiments. For example, if we consider a case, when exciting line-width $\Delta \omega$ is much larger than Doppler width $\Delta \omega_D$ of the atomic line (as it was originally assumed in the perturbation theory approach as given in [12])

$$\Delta \omega \gg \Delta \omega_D$$

then, as mentioned above, the atomic response does not depend on the velocity of translation motion of the atom and quantum density matrix $\rho$ refers to internal variables only. In such a case we obtain the rate equations by simply putting $k_\omega v = 0$ in (44), (45), which is the same as to consider the atomic velocity group $k_\omega v = 0$ only. Only one velocity group is also involved in experiments with cold atomic gases, atomic beams, etc.

However, we have successfully used the rate equations for Zeeman coherences (44), (45) in modeling of various experiments [18, 19, 20, 21]. In these experiments (59) clearly did not hold, nevertheless in describing experimental signal from all velocity groups, we have used the calculated signal from just one velocity group $k_\omega v = 0$ (note that $\Delta E_p (k_\omega v = 0) = 0$).

It is clear, that in this case for the experimental and simulation results to coincide, we cannot use the exact expressions (47), (49) for $\Gamma_p (k_\omega v = 0)$. Thus we must consider the "effective" induced transition rate $\Gamma_{eff}^p$, which in general does not coincide with $\Gamma_p (k_\omega v = 0)$.

Using the signal from velocity group $k_\omega v = 0$ as the calculated signal is justified, if we know the relation between $\Gamma_p (k_\omega v = 0)$ and $\Gamma_{eff}^p$ in advance. In reality this relation is
known only in some specific cases – for example, for the "steady-state" excitation with laser intensities below saturation – then we know, that $\Gamma^\text{eff}_p \sim \Gamma_p (k_\omega v = 0)$, as the signal from velocity group $k_\omega v = 0$ is proportional to the signal from all velocity groups – see [17]. However, in most cases establishing the relation between $\Gamma_p (k_\omega v = 0)$ and $\Gamma^\text{eff}_p$ is rather complicated, as it involves a large amount of calculations.

Therefore in analysis of experiments we have used the following approach – the signal from velocity group $k_\omega v = 0$ is calculated and then the best fit to an experiment is found – thus experimentally finding the relation between $\Gamma_p (k_\omega v = 0)$ and $\Gamma^\text{eff}_p$. In order to predict further results, we use the extrapolation and various other mathematical techniques. This method has proven to be successful in many cases.

C. Steady-state excitation.

As it was shown above generally, the usage of the rate equations for Zeeman coherences for description of time dependent behavior of atoms in laser and magnetic fields requires certain conditions regarding absorption rate connected with light intensity and spectral width of the laser line. At the same time very often in coherent atomic excitation experiments the "steady-state" or stationary excitation conditions are reached – the excitation light does not depend on time, which implies the same for the total density matrix $\rho(t)$. For the steady-state to happen, the system has to go over a rather large number of cycles, which means, that the steady-state is reached only after some time, after which it remains in this constant state forever (unless, of course, the conditions imposed on the system are changed). This means, that mathematically we can obtain the steady-state, if we consider $\rho(t = \infty)$ at some certain time moment in eternity $t = \infty$, when we can be sure, that the system has reached its steady-state - of course, if it can reach the steady-state at all. It is also obvious, that for steady-state the time derivative of the density matrix is zero and thus mathematically we can also obtain the steady-state by simply putting $d\rho(t)/dt = 0$ for both optical and Zeeman coherences.

Thus under the steady-state conditions we can express the optical coherences in terms of Zeeman coherences from the OBE straightforward, without any assumptions. In doing so we obtain the rate equations for Zeeman coherences $\rho_{g_i g_j}(t)$ and $\rho_{e_i e_j}(t)$, which now form a set of linear equations, because of the steady-state condition:

$$\begin{align*}
\frac{d\rho_{g_i g_j}(t)}{dt} &= 0, \\
\frac{d\rho_{e_i e_j}(t)}{dt} &= 0.
\end{align*}$$

As mentioned above, under the steady-state conditions in principle there is no limitations in the use of the rate equations, except the steady-state condition (60) itself.

D. The case of large Zeeman splitting.

In the case of large Zeeman splitting, that is, when (39) does not hold, the final rate equations become more complicated. However, the derivation procedure, of course, is still the same: we assume the already mentioned conditions, then simplify optical coherences $\rho_{g_i e_j}$ and $\rho_{e_i g_j}$ from (36), (37), and substitute them in (35), (38). Thus we arrive to the final
rate equations, which now become more complicated than (44), (45). The definitions of $\Gamma_p$ and $\Delta E_p$ also become different than those in (47) – (50). All the above analysis still holds.

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