# Validity of rate equations for Zeeman coherences for analysis of nonlinear interaction of atoms with broadband laser radiation

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In this paper we 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 in a straightforward way. We compare our approach with the perturbation-theory approach as given previously and show that our approach is more flexible for 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 the 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.

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## I. INTRODUCTION

Coherent effects in the interaction of laser radiation with atoms and molecules play a major role in physics and chemistry. Applications such as electromagnetically induced transparency [1], laser cooling [2–4], lasing without inversion [5], coherent population transfer [6], various nonlinear magneto-optical effects [7], new methods for magnetometry [8,9], coherent control of chemical reactions [10], and many others are widely used as 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, where direct solution of the time-dependent Schrödinger equation can be used [6], usually when one speaks about modeling of experiments with atomic coherent excitation, they mean the so-called "optical Bloch equations" (OBE) or Liouville equations [11–13] for the 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 subrecoil laser cooling mechanisms [2–4]. Optical coherences, on the other hand, are very sensitive to a variety of factors—collisions, finite laser linewidth, laser light fluctuations—both in phase and in amplitude, and many others. This means that, in describing a wide variety of atomic excitation experiments, one can neglect the optical coherences. This leads to the well-known *rate equations for Zeeman coherences* [12,14]. By saying "rate equations" we mean that these equations do not couple Zeeman coherences to optical coherences.

Among the first to obtain the rate equations for Zeeman coherences by neglecting the optical coherences were Cohen-Tannoudji and 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 a 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 the 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. The lack of rigorous mathematical argument was overcome later by 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 linewidth of the laser light used in excitation of atomic transition $\Delta\omega$ is very large compared to the natural linewidth $\Gamma$ of the atomic transition,

$$\Delta\omega \gg \Gamma, \tag{1}$$

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

$$\delta\omega < \Gamma. \tag{2}$$

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 the translational motion of the atom, and the quantum density matrix $\rho$ refers to internal variables only. In order to use the perturbation theory, the following condition must be satisfied:

$$\Delta\omega \gg \Gamma, \Gamma_p, \tag{3}$$

where $\Gamma_p$ is related to the time $T_p = 1/\Gamma_p$ characterizing the evolution of the density matrix $\rho$ under the effect of the coupling with the light beam. The rate equations for Zeeman coherences, obtained by considering the conditions (1)–(3), 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 seems to be substantially less demanding technically than the OBE from the viewpoint of implementation in the form of computer routines.

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

However, recently we have applied these equations for a description of some linear and nonlinear magneto-optical effects in stationary interaction of alkali-metal atoms with a broadband diode-laser radiation [18–21]. 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 first sight. For example, in 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 many coherent atomic excitation experiments.

What was the reason for such a good agreement between simulation and experiment in the above experiments with alkali-metal atoms? After a detailed analysis, it turned out that the key factor was the fact that the spectral linewidth of the radiation from the diode lasers was mainly determined by phase fluctuations. The problem was that rigorous analysis of the limitations of the rate equations for Zeeman coherences in the case of noisy laser radiation seemed still to be 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 linewidth 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 we 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 emphasis on the 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 good agreement with experiment that we feel that the relatively simple rate-equation approach (compared to the conventional OBE approach) is far too often undeservedly neglected when discussing the modeling of nonlinear magneto-optical effects in atoms.

## II. BROADBAND RADIATION INTERACTION WITH ATOMS

### A. Exciting light

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

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

$$\varepsilon(t) = |\varepsilon_{\bar{\omega}}| e^{-i\Phi(t) - i(\bar{\omega} - \mathbf{k}_{\bar{\omega}}\cdot\mathbf{v})t}. \tag{5}$$

We account for the shift $\bar{\omega} - \mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}$ in the laser frequency due to the Doppler effect: $\mathbf{v}$ is the velocity of translation motion of atoms and $\mathbf{k}_{\bar{\omega}}$ is the wave vector of the exciting light. $\bar{\omega}$ is the center frequency of the spectrum, $|\varepsilon_{\bar{\omega}}|$ 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 corresponds to a single-mode laser with a randomly fluctuating phase. In the case of a spectral lamp, this corresponds to a lamp where the dominant mechanism for the linewidth broadening is collisions between the radiating atoms or molecules. Note that for a single-mode laser, the BLA condition (2) is not fulfilled.

The Rabi frequency $\Omega_R$ is

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

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 the 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 a definite velocity $\mathbf{v}$, illuminated by the exciting light (4) and (5), resonant with the $g \leftrightarrow e$ transition, in the 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],

$$i\hbar\frac{\partial\rho}{\partial t} = [\tilde{H}, \tilde{\rho}] + i\hbar\hat{R}\rho, \tag{7}$$

we consider only the relaxation $\hat{R}$ 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 sim-

plicity, we also assume that the atomic transition forms a closed system—a cycling transition. In this spontaneous relaxation case, the terms for a closed system for the density matrix elements $\rho_{g_ig_j}$, $\rho_{g_ie_j}$, $\rho_{e_ig_j}$, $\rho_{e_ie_j}$ are

$$\hat{R}\rho_{g_ig_j} = \sum_{e_ie_j} \Gamma_{g_ig_j}^{e_ie_j}\rho_{e_ie_j},$$

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

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

$$\hat{R}\rho_{e_ie_j} = -\Gamma\rho_{e_ie_j}, \tag{8}$$

where $\Gamma_{g_ig_j}^{e_ie_j}$ describes the spontaneous relaxation from $\rho_{e_ie_j}$ to $\rho_{g_ig_j}$ and $\Gamma$ describes the spontaneous relaxation from $e$ to $g$. For the closed system it is obvious that

$$\sum_{g_ig_j} \Gamma_{g_ig_j}^{e_ie_j} = \Gamma.$$

The 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 $\hat{V}=-\hat{\mathbf{d}}\cdot\mathbf{E}(t)$, where $\hat{\mathbf{d}}$ is the electric dipole operator. Writing OBE explicity for the density matrix element $\rho_{ij}$, we get

$$\begin{aligned}\frac{\partial\rho_{ij}}{\partial t} &= -\frac{i}{\hbar}[\hat{H}_0,\rho_{ij}] + \frac{i}{\hbar}[\hat{\mathbf{d}}\cdot\mathbf{E}(t),\rho_{ij}] + \hat{R}\rho_{ij} \\ &= -i\omega_{ij}\rho_{ij} + \hat{R}\rho_{ij} + \sum_k\left(\frac{i}{\hbar}\varepsilon d_{ik}\rho_{kj} + \frac{i}{\hbar}\varepsilon^* d_{ik}^*\rho_{kj}\right. \\ &\quad \left. -\frac{i}{\hbar}\varepsilon d_{kj}\rho_{ik} - \frac{i}{\hbar}\varepsilon^* d_{kj}^*\rho_{ik}\right),\end{aligned} \tag{9}$$

where $\omega_{ij}=(E_i-E_j)/\hbar$ denotes the Zeeman splitting of the levels $i$ and $j$, and $d_{ik}\equiv\langle i|\mathbf{d}\cdot\mathbf{e}|k\rangle$, By choosing the quantization axis to be parallel to the external static magnetic field $\mathbf{B}$, all of the dependence of the density matrix on the $B$ field is included in the splitting term $\omega_{ij}$. Thus we arrive at the following equations for the density matrix elements $\rho_{g_ig_j}$, $\rho_{g_ie_j}$, $\rho_{e_ig_j}$, and $\rho_{e_ie_j}$:

$$\begin{aligned}\frac{\partial\rho_{g_ig_j}}{\partial t} &= \sum_{e_k}\left(\frac{i}{\hbar}\varepsilon d_{g_ie_k}\rho_{e_kg_j} + \frac{i}{\hbar}\varepsilon^* d_{g_ie_k}^*\rho_{e_kg_j} - \frac{i}{\hbar}\varepsilon d_{e_kg_j}\rho_{g_ie_k}\right. \\ &\quad \left. -\frac{i}{\hbar}\varepsilon^* d_{e_kg_j}^*\rho_{g_ie_k}\right) - i\omega_{g_ig_j}\rho_{g_ig_j} + \sum_{e_ie_j}\Gamma_{g_ig_j}^{e_ie_j}\rho_{e_ie_j},\end{aligned} \tag{10}$$

$$\begin{aligned}\frac{\partial\rho_{g_ie_j}}{\partial t} &= \sum_{e_k}\left(\frac{i}{\hbar}\varepsilon d_{g_ie_k}\rho_{e_ke_j} + \frac{i}{\hbar}\varepsilon^* d_{g_ie_k}^*\rho_{e_ke_j}\right) - \sum_{g_k}\left(\frac{i}{\hbar}\varepsilon d_{g_ke_j}\rho_{g_ig_k}\right. \\ &\quad \left. -\frac{i}{\hbar}\varepsilon^* d_{g_ke_j}^*\rho_{g_ig_k}\right) - i\omega_{g_ie_j}\rho_{g_ie_j} - \frac{\Gamma}{2}\rho_{g_ie_j},\end{aligned} \tag{11}$$

$$\begin{aligned}\frac{\partial\rho_{e_ig_j}}{\partial t} &= \sum_{g_k}\left(\frac{i}{\hbar}\varepsilon d_{e_ig_k}\rho_{g_kg_j} + \frac{i}{\hbar}\varepsilon^* d_{e_ig_k}^*\rho_{g_kg_j}\right) - \sum_{e_k}\left(\frac{i}{\hbar}\varepsilon d_{e_kg_j}\rho_{e_ie_k}\right. \\ &\quad \left. -\frac{i}{\hbar}\varepsilon^* d_{e_kg_j}^*\rho_{e_ie_k}\right) - i\omega_{e_ig_j}\rho_{e_ig_j} - \frac{\Gamma}{2}\rho_{e_ig_j},\end{aligned} \tag{12}$$

$$\begin{aligned}\frac{\partial\rho_{e_ie_j}}{\partial t} &= \sum_{g_k}\left(\frac{i}{\hbar}\varepsilon d_{e_ig_k}\rho_{g_ke_j} + \frac{i}{\hbar}\varepsilon^* d_{e_ig_k}^*\rho_{g_ke_j} - \frac{i}{\hbar}\varepsilon d_{g_ke_j}\rho_{e_ig_k}\right. \\ &\quad \left. -\frac{i}{\hbar}\varepsilon^* d_{g_ke_j}^*\rho_{e_ig_k}\right) - i\omega_{e_ie_j}\rho_{e_ie_j} - \Gamma\rho_{e_ie_j}.\end{aligned} \tag{13}$$

The matrix elements of the type $d_{e_ig_j}\equiv\langle e_i|\mathbf{d}\cdot\mathbf{e}|g_j\rangle$ 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:

$$\begin{aligned}\rho_{g_ig_j} &= \widetilde{\rho_{g_ig_j}} = \rho_{g_ig_j}, \\ \rho_{g_ie_j} &= \widetilde{\rho_{g_ie_j}}e^{i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v})t+i\Phi(t)}, \\ \rho_{e_ig_j} &= \widetilde{\rho_{e_ig_j}}e^{-i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v})t-i\Phi(t)}, \\ \rho_{e_ie_j} &= \widetilde{\rho_{e_ie_j}} = \rho_{e_ie_j}.\end{aligned} \tag{14}$$

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

$$\begin{aligned}\frac{\partial\rho_{g_ig_j}}{\partial t} &= \sum_{e_k}\left(\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d_{g_ie_k}^*\widetilde{\rho_{e_kg_j}} - \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d_{e_kg_j}\widetilde{\rho_{g_ie_k}}\right) - i\omega_{g_ig_j}\rho_{g_ig_j} \\ &\quad + \sum_{e_ie_j}\Gamma_{g_ig_j}^{e_ie_j}\rho_{e_ie_j},\end{aligned} \tag{15}$$

$$\begin{aligned}\frac{\partial\widetilde{\rho_{g_ie_j}}}{\partial t} &= \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{e_k} d_{g_ie_k}^*\rho_{e_ke_j} - \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{g_k} d_{g_ke_j}^*\rho_{g_ig_k} \\ &\quad - i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\mathbf{v}+\omega_{g_ie_j})\widetilde{\rho_{g_ie_j}} - \frac{\Gamma}{2}\widetilde{\rho_{g_ie_j}} - i\frac{\partial\Phi(t)}{\partial t}\widetilde{\rho_{g_ie_j}},\end{aligned} \tag{16}$$

$$\begin{aligned}\frac{\partial\widetilde{\rho_{e_ig_j}}}{\partial t} &= \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{g_k} d_{e_ig_k}\rho_{g_kg_j} - \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{e_k} d_{e_kg_j}\rho_{e_ie_k} \\ &\quad + i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\mathbf{v}-\omega_{e_ig_j})\widetilde{\rho_{e_ig_j}} - \frac{\Gamma}{2}\widetilde{\rho_{e_ig_j}} + i\frac{\partial\Phi(t)}{\partial t}\widetilde{\rho_{e_ig_j}},\end{aligned} \tag{17}$$

$$\begin{aligned}\frac{\partial\rho_{e_ie_j}}{\partial t} &= \sum_{g_k}\left(\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d_{e_ig_k}\widetilde{\rho_{g_ke_j}} - \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d_{g_ke_j}^*\widetilde{\rho_{e_ig_k}}\right) - i\omega_{e_ie_j}\rho_{e_ie_j} \\ &\quad - \Gamma\rho_{e_ie_j}.\end{aligned} \tag{18}$$

### C. Atoms in a fluctuating optical field

Equations (15)–(18) are stochastic differential equations [27] with stochastic variable $\partial\Phi(t)/\partial t$. In an experiment, as a rule, we deal with quantities that are averaged over the time intervals that are large in comparison with the phase-fluctuation time in the excitation-light source, therefore we need to perform a statistical averaging of the above equations. In order to do that, we solve Eqs. (16) and (17) [with initial condition $\rho_{g_ie_j}(t_0)=\rho_{e_ig_j}(t_0)=0$] and then take a formal statistical average over the fluctuating phases,

$$\frac{\partial\langle\rho_{g_ig_j}\rangle}{\partial t}=\sum_{e_k}\left(\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d^*_{g_ie_k}\widetilde{\langle\rho_{e_kg_j}\rangle}-\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d_{e_kg_j}\widetilde{\langle\rho_{g_ie_k}\rangle}\right)-i\omega_{g_ig_j}\langle\rho_{g_ig_j}\rangle+\sum_{e_ie_j}\Gamma^{e_ie_j}_{g_ig_j}\langle\rho_{e_ie_j}\rangle, \tag{19}$$

$$\begin{aligned}\widetilde{\langle\rho_{g_ie_j}\rangle}=&\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{e_k}d^*_{g_ie_k}\int_{t_0}^{t}e^{[-i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\mathbf{v}+\omega_{g_ie_j})-(\Gamma/2)](t-t')}\\&\times\langle\rho_{e_ke_j}(t')e^{-i[\Phi(t)-\Phi(t')]}\rangle dt'\\&-\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{g_k}d^*_{g_ke_j}\int_{t_0}^{t}e^{[-i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\mathbf{v}+\omega_{g_ie_j})-(\Gamma/2)](t-t')}\\&\times\langle\rho_{g_ig_k}(t')e^{-i[\Phi(t)-\Phi(t')]}\rangle dt',\end{aligned} \tag{20}$$

$$\begin{aligned}\widetilde{\langle\rho_{e_ig_j}\rangle}=&\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{g_k}d_{e_ig_k}\int_{t_0}^{t}e^{[i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\mathbf{v}-\omega_{e_ig_j})-(\Gamma/2)](t-t')}\\&\times\langle\rho_{g_kg_j}(t')e^{i[\Phi(t)-\Phi(t')]}\rangle dt'\\&-\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{e_k}d_{e_kg_j}\int_{t_0}^{t}e^{[i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\mathbf{v}-\omega_{e_ig_j})-(\Gamma/2)](t-t')}\\&\times\langle\rho_{e_ie_k}(t')e^{i[\Phi(t)-\Phi(t')]}\rangle dt',\end{aligned} \tag{21}$$

$$\frac{\partial\langle\rho_{e_ie_j}\rangle}{\partial t}=\sum_{g_k}\left(\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d^*_{e_ig_k}\widetilde{\langle\rho_{g_ke_j}\rangle}-\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d^*_{g_ke_j}\widetilde{\langle\rho_{e_ig_k}\rangle}\right)-i\omega_{e_ie_j}\langle\rho_{e_ie_j}\rangle-\Gamma\langle\rho_{e_ie_j}\rangle. \tag{22}$$

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

$$\langle\rho_{a_ia_j}(t')e^{\pm i[\Phi(t)-\Phi(t')]}\rangle=\langle\rho_{a_ia_j}(t')\rangle\langle e^{\pm i[\Phi(t)-\Phi(t')]}\rangle, \tag{23}$$

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 [32–34], which assumes that the phase remains constant except for sudden random "jump," when it changes to a new constant value. This model is used to describe the fluctuations from spectral lamps when the dominant mechanism for the linewidth 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 single-mode lasers 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 the case of optical Bloch equations performed in [32–34]. The random jump process is Poissonian in nature—the probability for the phase to change $N$ times during time $t-t'$ is

$$P_N=\frac{1}{N!}\left(\frac{t-t'}{T}\right)^N e^{-(t-t')/T}, \tag{24}$$

where $T$ is the average time between successive phase jumps. Now we define $\langle e^{\pm i\Delta\Phi}\rangle$ 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 on 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 $N=0-\infty$ of phase jumps during time $t-t'$,

$$\begin{aligned}\langle e^{\pm i[\Phi(t)-\Phi(t')]}\rangle&=\sum_{N=0}^{\infty}P_N\langle e^{\pm i[\Phi(t)-\Phi(t')]}\rangle_N\\&=e^{-(t-t')/T}\sum_{N=0}^{\infty}\frac{1}{N!}\left(\frac{t-t'}{T}\langle e^{\pm i\Delta\Phi}\rangle\right)^N\\&=e^{[(t-t')/T](1-\langle e^{\pm i\Delta\Phi}\rangle)}.\end{aligned} \tag{25}$$

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 (on average after the time $T$ the phase "forgets" its past) $T=2/\Delta\omega$. We also assume that all phase values occur with equal probability. Then $\langle e^{\pm i\Delta\Phi}\rangle=0$, and Eq. (25) becomes

$$\langle e^{\pm i[\Phi(t)-\Phi(t')]}\rangle=e^{-(\Delta\omega/2)(t-t')}. \tag{26}$$

This means that the spectral distribution of the exciting light is Lorentzian with a 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 is based on the phase-diffusion model analyzed in [29–31,35,36]. In the phase-diffusion model, the field has a constant amplitude, but its phase is a fluctuating quantity which obeys the Langevin equation

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

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

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

$$\langle \varsigma(t)\rangle = 0, \tag{29}$$

which means that $\varsigma(t)$ obeys the Langevin equation for Brownian motion [22,27],

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

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

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

The meaning of the parameters $b$ and $\beta$ can be understood from Eqs. (27)–(31). The quantity $1/\beta$ is the correlation time of the phase time derivative $\varsigma(t)$, but $b$ gives the bandwidth 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 Eqs. (27)–(31) is given by the Fourier transform of the correlation function,

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

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

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

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

but the phase $\Phi(t)$ determined by 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]. It is easily understood as in this process the relevant fluctuating quantity $\varsigma(t)$ is $\delta$-correlated [the correlation time $1/\beta$ of $\varsigma(t)$ tends to zero when $\beta\to\infty$], and thus we can always separate the time scales of evolution of $\langle \rho_{a_i a_j}(t')\rangle$ and $\langle e^{\pm i[\Phi(t)-\Phi(t')]}\rangle$ in Eq. (23). The 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 the Wiener-Levy process, the relation (32) becomes

$$\langle e^{\pm i[\Phi(t)-\Phi(t')]}\rangle = \exp[-b|t-t'|] = e^{-(\Delta\omega/2)(t-t')}, \tag{34}$$

where we have used the fact that $t \geqslant t'$.

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 a cutoff at frequencies $\beta$. In this case, the damping term $\Delta\omega/2$ is simply multiplied by the cutoff 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 $\varsigma(t) = d\Phi(t)/dt$.

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

As can be seen [Eqs. (26) and (34)], both approaches give similar results for the time-averaged 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 Eqs. (26) and (34) to rewrite Eqs. (19)–(22) (for simplicity in the subsequent expressions, we further drop the averaging brackets),

$$\frac{\partial \rho_{g_i g_j}}{\partial t} = \sum_{e_k}\left(\frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d^*_{g_i e_k}\widetilde{\rho_{e_k g_j}} - \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d_{e_k g_j}\widetilde{\rho}_{g_i e_k}\right) - i\omega_{g_i g_j}\rho_{g_i g_j} + \sum_{e_i e_j}\Gamma^{e_i e_j}_{g_i g_j}\rho_{e_i e_j}, \tag{35}$$

$$\widetilde{\rho_{g_i e_j}} = \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{e_k} d^*_{g_i e_k}\int_{t_0}^{t}\exp[-i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}+\omega_{g_i e_j}) - (\Gamma/2+\Delta\omega/2)](t-t')\rho_{e_k e_j}(t')dt' - \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{g_k} d^*_{g_k e_j}\int_{t_0}^{t}\exp[-i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}+\omega_{g_i e_j}) - (\Gamma/2+\Delta\omega/2)](t-t')\rho_{g_i g_k}(t')dt', \tag{36}$$

$$\widetilde{\rho_{e_i g_j}} = \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{g_k} d_{e_i g_k}\int_{t_0}^{t}\exp[i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}-\omega_{e_i g_j}) - (\Gamma/2+\Delta\omega/2)](t-t')\rho_{g_k g_j}(t')dt' - \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|\sum_{e_k} d_{e_k g_j}\int_{t_0}^{t}\exp[i(\bar{\omega}-\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}-\omega_{e_i g_j}) - (\Gamma/2+\Delta\omega/2)](t-t')\rho_{e_i e_k}(t')dt', \tag{37}$$

$$\frac{\partial \rho_{e_i e_j}}{\partial t} = \sum_{g_k}\left(\frac{1}{\hbar}|\varepsilon_{\bar{\omega}}|d_{e_i g_k}\widetilde{\rho_{g_k e_j}} - \frac{i}{\hbar}|\varepsilon_{\bar{\omega}}|d^*_{g_k e_j}\widetilde{\rho_{e_i g_k}}\right) - i\omega_{e_i e_j}\rho_{e_i e_j} - \Gamma\rho_{e_i e_j}. \tag{38}$$

## III. RATE EQUATIONS

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

$$\Delta\omega \gg \omega_B, \tag{39}$$

though this condition can be avoided at the expense of complicating the final rate equations. Equation (39) means that we can write

$$\left(\frac{\Gamma}{2}+\frac{\Delta\omega}{2}\right)+i(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}+\omega_{g_ie_j}) \approx \left(\frac{\Gamma}{2}+\frac{\Delta\omega}{2}\right)+i(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0), \tag{40}$$

$$\left(\frac{\Gamma}{2}+\frac{\Delta\omega}{2}\right)-i(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}+\omega_{e_ig_j}) \approx \left(\frac{\Gamma}{2}+\frac{\Delta\omega}{2}\right)+i(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0), \tag{41}$$

At this point we go further and assume certain conditions which allow us to simplify significantly the expressions for optical coherences (36) and (37). These conditions are either BLA (1)–(3) or the steady-state [(60)—see below] conditions. Under these circumstances, the expressions for optical coherences (36) and (37) become

$$\widetilde{\rho_{g_ie_j}}=\frac{i}{\hbar}\frac{|\varepsilon_{\overline{\omega}}|}{\left(\frac{\Gamma}{2}+\frac{\Delta\omega}{2}\right)+i(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\mathbf{v}-\omega_0)} \times\left(\sum_{e_k} d^*_{g_ie_k}\rho_{e_ke_j}-\sum_{g_k} d^*_{g_ke_j}\rho_{g_ig_k}\right), \tag{42}$$

$$\widetilde{\rho_{e_ig_j}}=\frac{i}{\hbar}\frac{|\varepsilon_{\overline{\omega}}|}{\left(\frac{\Gamma}{2}+\frac{\Delta\omega}{2}\right)-i(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\mathbf{v}-\omega_0)} \times\left(\sum_{g_k} d_{e_ig_k}\rho_{g_kg_j}-\sum_{e_k} d_{e_kg_j}\rho_{e_ie_k}\right). \tag{43}$$

Now, by substituting Eqs. (42) and (43) in Eqs. (35) and (38), we arrive at the final rate equations,

$$\frac{\partial\rho_{g_ig_j}}{\partial t}=\Gamma_p\sum_{e_k,e_m}(d_1^{g_ie_k})^*d_1^{e_mg_j}\rho_{e_ke_m}-\sum_{e_k,g_m}\left[\left(\frac{\Gamma_p}{2}+i\Delta E_p\right)\times(d_1^{g_ie_k})^*d_1^{e_kg_m}\rho_{g_mg_j}-\left(\frac{\Gamma_p}{2}-i\Delta E_p\right)(d_1^{g_me_k})^*\times d_1^{e_kg_j}\rho_{g_ig_m}\right]-i\omega_{g_ig_j}\rho_{g_ig_j}+\sum_{e_ie_j}\Gamma^{e_ie_j}_{g_ig_j}\rho_{e_ie_j}, \tag{44}$$

$$\frac{\partial\rho_{e_ie_j}}{\partial t}=\Gamma_p\sum_{g_k,g_m} d_1^{e_ig_k}(d_1^{g_me_j})^*\rho_{g_kg_m}-\sum_{g_k,e_m}\left[\left(\frac{\Gamma_p}{2}-i\Delta E_p\right)\times d_1^{e_ig_k}\left(d_1^{g_ke_m}\right)^*\rho_{e_me_j}-\left(\frac{\Gamma_p}{2}+i\Delta E_p\right)\times d_1^{e_mg_k}(d_1^{g_ke_j})^*\rho_{e_ie_m}\right]-i\omega_{e_ie_j}\rho_{e_ie_j}-\Gamma\rho_{e_ie_j}, \tag{45}$$

where $d_1^{e_mg_j}\equiv\langle e_m|\mathbf{d}_1\cdot\mathbf{e}|g_j\rangle$ and $\mathbf{d}_1$ denotes the electric dipole moment unity vector ($\mathbf{d}_1=\mathbf{d}/|d|$), and thus matrix elements $d_{e_ig_j}$ are written as

$$d_{e_ig_j}=d_1^{e_ig_j}\langle e\|d\|g\rangle, \tag{46}$$

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

$$\frac{\Gamma_p}{2}=\frac{|\varepsilon_{\overline{\omega}}|^2}{\hbar^2}\times|\langle e\|d\|g\rangle|^2\times\frac{\left(\frac{\Gamma}{2}+\frac{\Delta\omega}{2}\right)}{\left(\frac{\Gamma}{2}+\frac{\Delta\omega}{2}\right)^2+(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0)^2}, \tag{47}$$

$$\Delta E_p=\frac{|\varepsilon_{\overline{\omega}}|^2}{\hbar^2}\times|\langle e\|d\|g\rangle|^2\times\frac{\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0}{\left(\frac{\Gamma}{2}+\frac{\Delta\omega}{2}\right)^2+(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0)^2}. \tag{48}$$

The quantity $\Gamma_p$ is the probability per unit time of an absorption of 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), Eqs. (47) and (48) become

$$\frac{\Gamma_p}{2}\approx\frac{|\varepsilon_{\overline{\omega}}|^2}{\hbar^2}\times|\langle e\|d\|g\rangle|^2\times\frac{\frac{\Delta\omega}{2}}{\left(\frac{\Delta\omega}{2}\right)^2+(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0)^2}, \tag{49}$$

$$\Delta E_p\approx\frac{|\varepsilon_{\overline{\omega}}|^2}{\hbar^2}\times|\langle e\|d\|g\rangle|^2\times\frac{\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0}{\left(\frac{\Delta\omega}{2}\right)^2+(\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0)^2}. \tag{50}$$

Note also that the phase fluctuations (described by the above models) reduce the saturation on resonance ($\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0=0$) by a factor $\Gamma/(\Gamma+\Delta\omega)$, and increase the saturation faroff resonance ($\overline{\omega}-\mathbf{k}_{\overline{\omega}}\cdot\mathbf{v}-\omega_0\gg\Gamma,\Delta\omega$) by a factor $(\Gamma+\Delta\omega)/\Gamma$.

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

$$I(\vec{e}_1)=\widetilde{I}_0\sum_{g_i,e_i,e_j}(d^{(ob)}_{g_ie_j})^*d^{(ob)}_{e_ig_i}\rho_{e_ie_j}, \tag{51}$$

where $\widetilde{I}_0$ is a proportionality coefficient and the matrix element $d^{(ob)}_{g_ie_j}=\langle g_i|\mathbf{d}\cdot\mathbf{e}_1|e_j\rangle$ contains the polarization vector $\mathbf{e}_1$ of the light which is detected.

## IV. ANALYSIS AND CONCLUSIONS

### A. Perturbation theory approach

The obtained rate equations for Zeeman coherences coincide with equations obtained earlier in the perturbation theory approach in [12]. In that approach the ratios between rate constants involved in the problem $(\Gamma_p,\Gamma)$ and the linewidth of the excitation radiation $\Delta\omega$ are used as small parameters. Here we stress that, although 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 the two approaches, let us have a brief look at the method used and conclusions obtained with perturbation theory.

Let $T_p=1/\Gamma_p$ be the time characterizing the evolution of the density matrix under the effect of coupling with the light field. In the following analysis, it is assumed that the intensity is sufficiently low so that $T_p$ is much longer than the correlation time $T=1/\Delta\omega$ of the light wave,

$$\Delta\omega \gg \Gamma_p. \tag{52}$$

Now consider a time interval $\Delta t$ such that

$$T_p,\tau \gg \Delta t \gg T, \tag{53}$$

where $\tau=1/\Gamma$. 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 $\rho(t)$,

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

This means that we can replace $\Delta\rho(t)/\Delta t$ with the time derivative $d\rho(t)/dt$, provided that we never use $d\rho(t)/dt$ 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. The quantity $\Delta\rho(t)/\Delta t=d\rho(t)/dt$ is called the "coarse-grained" derivative [40].

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

$$\mathbf{E}(t)=\varepsilon(t)\mathbf{e}+\varepsilon^*(t)\mathbf{e}^*, \tag{55}$$

$$\varepsilon(t)=\sum_\mu |\varepsilon_\mu| e^{-i\Phi_\mu - i(\omega_\mu - \mathbf{k}\omega_\mu\cdot\mathbf{v})t}. \tag{56}$$

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

$$\langle e^{-i(\Phi_\mu-\Phi_{\mu'})}\rangle=\delta_{\mu\mu'}. \tag{57}$$

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

$$\begin{aligned}\langle\varepsilon(t)\varepsilon^*(t-\tau)\rangle &= \sum_{\mu,\mu'} |\varepsilon_\mu||\varepsilon_{\mu'}|\langle e^{-i(\Phi_\mu-\Phi_{\mu'})}\rangle \\ &\quad\times e^{-i(\omega_\mu-\mathbf{k}\omega_\mu\cdot\mathbf{v})t} e^{i(\omega_{\mu'}-\mathbf{k}\omega_\mu\cdot\mathbf{v})(t-\tau)} \\ &= \sum_\mu |\varepsilon_\mu|^2 e^{-i(\omega_\mu-\mathbf{k}\omega_\mu\cdot\mathbf{v})\tau}.\end{aligned} \tag{58}$$

Applying perturbation theory, after some calculations with the consideration of Eq. (58) and the "coarse-grained" derivative, the rate equations are obtained [12], again considering the condition (39) for simplicity. The obtained rate equations are exactly the same as the above derived equations (44) and (45), but with $\Gamma_p$ and $\Delta E_p$ having a slightly different form as defined in Eqs. (47)–(50). This mismatch is easily avoided if instead of the exciting-light model (55)–(58) we take the model (4), (5), (26), and (34). Then the rate equations are the same as Eqs. (44) and (45), with $\Gamma_p$ and $\Delta E_p$ defined as in Eqs. (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 at the conclusion stated above that the approach discussed in this paper allows us to examine the limits of usage of the rate equations for Zeeman coherences to a greater detail than the perturbation theory approach. Therefore, it can be applied to analyze a larger number or experimental situations.

### B. Velocity dependence

When we look at Eqs. (47)–(50), we see that $\Gamma_p$ (the induced transition rate) and $\Delta E_p$ (the dynamic Stark shift) are velocity-dependent, and thus also are Eqs. (44) and (45). This means that in describing the observable signal, we need to take into account all velocity groups involved (note that we have already assumed that different velocity groups do not interact—the density of atoms is sufficiently low). In a 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 Eqs. (44)–(50). Thus a large amount of calculations 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 where the exciting linewidth $\Delta\omega$ is much larger than the Doppler width $\Delta\omega_D$ of the atomic line (as it was originally assumed in the perturbation-theory approach as given in [12]),

$$\Delta\omega \gg \Delta\omega_D, \tag{59}$$

then, as mentioned above, the atomic response does not depend on the velocity of translation motion of the atom and the quantum density matrix $\rho$ refers to internal variables only. In such a case, we obtain the rate equations by simply

putting $k_{\varpi}v=0$ in Eqs. (44) and (45), which is the same as considering the atomic velocity group $k_{\varpi}v=0$ only. Only one velocity group is also involved in experiments with cold atomic gases, atomic beams, etc.

Nevertheless, we have successfully used the rate equations for Zeeman coherences (44) and (45) in modeling various experiments [18–21]. In these experiments, Eq. (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 $k_{\bar{\omega}}v=0$ [note that at resonance $\Delta E_p(\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}=0)=0$]. It is clear, that in this case for the experimental and simulation results to coincide, we cannot use the exact expressions (47) and (49) for $\Gamma_p(\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}=0)$. Thus we must consider the "effective" induced transition rate $\Gamma_p^{\mathrm{eff}}$, which in general does not coincide with $\Gamma_p(\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}=0)$.

Using the signal from the velocity group $k_{\bar{\omega}}v=0$ as the calculated signal is justified if we know the relation between $\Gamma_p(\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}=0)$ and $\Gamma_p^{\mathrm{eff}}$ 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—so we know that $\Gamma_p^{\mathrm{eff}}\sim\Gamma_p(\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}=0)$, as the signal from the velocity group $k_{\bar{\omega}}v=0$ is proportional to the signal from all velocity groups—see [17]. However, in most cases establishing the relation between $\Gamma_p(\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}=0)$ and $\Gamma_p^{\mathrm{eff}}$ is rather complicated as it involves a large amount of calculations.

Therefore, in the analysis of experiments we have used the following approach—the signal from the velocity group $k_{\bar{\omega}}v=0$ is calculated and then the best fit to an experiment is found—thus experimentally finding the relation between $\Gamma_p(\mathbf{k}_{\bar{\omega}}\cdot\mathbf{v}=0)$ and $\Gamma_p^{\mathrm{eff}}$. 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 was shown above generally, the use of the rate equations for Zeeman coherences to describe the time-dependent behavior of atoms in laser and magnetic fields requires certain conditions regarding absorption rate related to the 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 $\rho(t)$. When an atom is suddenly placed in an optical field, the steady-state condition 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 solution if we consider $\rho(t=\infty)$. It is also obvious that for the 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 directly, without any assumptions. In doing so, we obtain the rate equations for Zeeman coherences $\rho_{g_ig_j}(t)$ and $\rho_{e_ie_j}(t)$, which now form a set of linear equations, because of the steady-state condition,

$$\frac{d\rho_{g_ig_j}(t)}{dt}=0,$$

$$\frac{d\rho_{e_ie_j}(t)}{dt}=0. \tag{60}$$

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

### D. The case of large Zeeman splitting

In the case of large Zeeman splittings, that is, when Eq. (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 $\widetilde{\rho_{g_ie_j}}$ and $\widetilde{\rho_{e_ig_j}}$ from Eqs. (36) and (37), and substitute them in Eqs. (35) and (38). Thus we arrive at the final rate equations, which now become more complicated than Eqs. (44) and (45). The definitions of $\Gamma_p$ and $\Delta E_p$ also become different from those in Eqs. (47)–(50). All of the above analysis still holds.

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[1] S. Harris, Phys. Today **50** (7), 36 (1997).
[2] S. Chu, Rev. Mod. Phys. **70**, 685 (1998).
[3] C. Cohen-Tannoudji, Rev. Mod. Phys. **70**, 707 (1998).
[4] W. D. Phillips, Rev. Mod. Phys. **70**, 721 (1998).
[5] M. Scully, S-Y. Zhu, and A. Gavrielides, Phys. Rev. Lett. **62**, 2813 (1989).
[6] K. Bergmann, H. Theuer, and B. W. Shore, Rev. Mod. Phys. **70**, 1003 (1998).
[7] D. Budker, W. Gawlik, D. F. Kimball, S. M. Rochester, and V. V. Yashchuk, Rev. Mod. Phys. **74**, 1153 (2002).
[8] M. Scully and M. Fleischhauer, Phys. Rev. Lett. **69**, 1360 (1992).
[9] D. Budker, Nature (London) **422**, 574 (2003).
[10] P. W. Brumer and M. Shapiro, *Principles of the Quantum Control of Molecular Processes* (Wiley-Interscience, Hoboken, NJ, 2003).
[11] L. Allen and J. H. Eberly, *Optical Resonance and Two Level Atoms* (Wiley, New York, 1975).

[12] C. Cohen-Tannoudji, in *Frontiers in Laser Spectroscopy* (North-Holland, Amsterdam, 1977).
[13] E. Arimondo, Prog. Opt. **35**, 257 (1996).
[14] J. P. Barrat and C. Cohen-Tannoudji, J. Phys. Radium **22**, 329 (1961); **22**, 443 (1961).
[15] M. Ducloy, Phys. Rev. A **8**, 1844 (1973).
[16] M. P. Auzinsh and R. S. Ferber, Sov. Phys. Usp. **33**, 833 (1990).
[17] M. Auzinsh and R. Ferber, *Optical Polarization of Molecules* (Cambridge University Press, Cambridge, 1995).
[18] J. Alnis and M. Auzinsh, Phys. Rev. A **63**, 023407 (2001).
[19] J. Alnis and M. Auzinsh, J. Phys. B **34**, 3889 (2001).
[20] J. Alnis, K. Blushs, M. Auzinsh, S. Kennedy, N. Shafer-Ray, and E. R.I. Abraham, J. Phys. B **36**, 1161 (2003).
[21] A. V. Papojan, M. Auzinsh, and K. Bergmann, Eur. Phys. J. D **21**, 63 (2002).
[22] S. Stenholm, *Foundations of Laser Spectroscopy* (Wiley, New York, 1984).
[23] L. P. Yatsenko, V. I. Romanenko, B. W. Shore, and K. Bergmann, Phys. Rev. A **65**, 043409 (2002).
[24] M. P. Auzinsh and R. S. Ferber, Phys. Rev. A **43**, 2374 (1991).
[25] D. A. Varshalovich, A. N. Moskalev, and V. K. Khersonskii, *Quantum Theory of Angular Momentum* (World Scientific, Singapore, 1988).
[26] R. N. Zare, *Angular Momentum, Understanding Spatial Aspects in Chemistry and Physics* (Wiley, New York, 1988).
[27] N. G. van Kampen, Phys. Lett., C **24**, 171 (1976).
[28] J. H. Eberly, Phys. Rev. Lett. **37**, 1387 (1976).
[29] A. T. Georges and P. Lambropoulos, Phys. Rev. A **18**, 587 (1978).
[30] A. T. Georges and P. Lambropoulos, Phys. Rev. A **20**, 991 (1979).
[31] A. T. Georges, Phys. Rev. A **21**, 2034 (1980).
[32] A. I. Burshtein, Sov. Phys. JETP **22**, 939 (1966).
[33] A. I. Burshtein and Yu. S. Oseledchik, Sov. Phys. JETP **24**, 716 (1967).
[34] J. H. Eberly, K. Wodkiewicz, and B. W. Shore, Phys. Rev. A **30**, 2381 (1984); Phys. Rev. A **30**, 2390 (1984).
[35] P. Zoller and P. Lambropoulos, J. Phys. B **12**, L547 (1979).
[36] S. N. Dixit, P. Zoller, and P. Lambropoulos, Phys. Rev. A **21**, 1289 (1980).
[37] H. Haken, in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1970), Vol. XXV/2c.
[38] P. Avan and C. Cohen-Tannoudji, J. Phys. B **10**, 155 (1977).
[39] M. I. Dyakonov, Sov. Phys. JETP **20**, 1484 (1965).
[40] K. Blum, *Density Matrix: Theory and Applications*, 2nd ed. (Plenum, New York, 1996).