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Title: Spin-degenerate two-level atoms in on-resonance partially polarized light

Year: 2006

Version: Final published version

Please cite the original version:

Shevchenko, Andriy & Kaivola, Matti & Javanainen, Juha. 2006. Spin-degenerate two-level atoms in on-resonance partially polarized light. Physical Review A. Volume 73, Issue 3, 035801. ISSN 1094-1622 (electronic). ISSN 1050-2947 (printed). DOI: 10.1103/physreva.73.035801.

PHYSICAL REVIEW A **73**, 035801 (2006)

Spin-degenerate two-level atoms in on-resonance partially polarized light

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(Received 26 August 2005; revised manuscript received 2 December 2005; published 23 March 2006)

We present a theoretical model describing the magnetic-state population dynamics of spin-degenerate two-level atoms interacting with a narrowband, on-resonance, partially polarized electromagnetic field. The field is allowed to have three uncorrelated orthogonal vector components. The model is applied to a four-magnetic-state atom system with a single excited and three ground states. Even if the field is narrowband, the population dynamics may be completely predicated by the fluctuating polarization of light. In our examples, the fluctuation effects are mainly governed by a single parameter, the degree of polarization of the field.

DOI: 10.1103/PhysRevA.73.035801 PACS number(s): 42.50.Ct, 32.80.Bx, 32.80.Qk, 42.25.Ja

When treating the interaction of atomic systems with electromagnetic fields, the fields are typically assumed to be planar. The fluctuations of the phase and amplitude are usually ignored, and if not, then the magnetic degeneracy of the atomic energy levels is not considered. In general, however, the atomic transitions take place between magnetic substates, and in order to obtain a complete picture of the atom-field interaction, one should note the general correspondence between the three basic orthogonal components of the transition dipole moment and the three possible orthogonal vector components of the field. These vector components can, and will, undergo fluctuations in a realistic situation. The resulting fluctuations of the field polarization will obviously affect the evolution of the magnetic-state populations and influence processes caused by quantum interference, such as population trapping in dark states [1–6].

We present a description of the interaction of spin-degenerate two-level atoms with a narrowband on-resonance field which is allowed to have all three orthogonal vector components with fluctuating phases. The phase fluctuations are described by applying the Ornstein-Uhlenbeck phase-diffusion model. The bandwidth of the field, B, is assumed to be smaller than the spontaneous emission rate Γ. Such fields, produced by realistic amplitude-stabilized lasers [7–10], are frequently used, e.g., in laser cooling and trapping experiments. Solving the evolution equations for the density-matrix elements, we study the dynamics of the magnetic-state populations. The main findings are two. First, no matter how narrowband the fluctuations are, they may determine the qualitative character of the dynamics of the populations. Second, in our examples a single parameter, the degree of polarization of the field, appears to suffice for the characterization of the fluctuating light—no detailed statistics is needed.

An arbitrary electromagnetic field can be described in terms of its three orthogonal vector components as $\vec{E}=E_x\vec{e}_x+E_y\vec{e}_y+E_z\vec{e}_z$. In a spherical-basis representation, $\{\vec{e}_{-1}=(\vec{e}_x-i\vec{e}_y)/\sqrt{2},\vec{e}_0=\vec{e}_z,\vec{e}_{+1}=-(\vec{e}_x+i\vec{e}_y)/\sqrt{2}\}$, with $\vec{z}$ being the quantization axis for the atoms, the field is expressed as $\vec{E}=E_{+1}\vec{e}^{\,*}_{-1}+E_0\vec{e}^{\,*}_0+E_{-1}\vec{e}^{\,*}_{+1}$ [1], with the components E_{-1},E_0, and E_{+1} driving the σ^-, π, and σ^+ transitions, respectively (the asterisk stands for complex conjugate). While analyzing the response of an atom to an electromagnetic field, the convention is to use spherical-basis representation of the vector quantities. We consider an atom with two energy levels which are angular-momentum degenerate. The magnetic states of the ground (excited) level are defined as $|\psi_j\rangle$ ($|\widetilde{\psi}_k\rangle$) with the subindices running through the magnetic quantum numbers m_F ($\widetilde{m}_F$); total angular momenta for the levels are F and $\widetilde{F}$. The vector components of the field, each driving transitions between states j and k, are written as $E_{k-j}=\mathcal{E}_{k-j}(\vec{r},t)\cos[w_r t+\varphi_{k-j}(\vec{r},t)]$, where $\varphi_{k-j}(\vec{r},t)$ is the fluctuating phase. We split up the density operator into ground-state and excited-state parts with the Zeeman components $\rho^g_{j,k}$ and $\rho^e_{j,k}$ describing populations when $j=k$ and coherences when $j\neq k$, and into a part $\rho_{j,k}$ that conveys coherences between Zeeman states in the upper and lower levels. A conventional calculation then gives the equations of motion for the density-matrix elements in the rotating frame as

$$\dot{\rho}^e_{j,k}=\frac{i}{2}\sum_{m=j-1}^{j+1}\rho_{m,k}\Omega_{m,j}e^{i\varphi_{j-m}}-\frac{i}{2}\sum_{n=k-1}^{k+1}\widetilde{\rho}_{j,n}\Omega_{n,k}e^{-i\varphi_{k-n}}-\Gamma\rho^e_{j,k}, \tag{1}$$

$$\dot{\rho}^g_{j,k}=\frac{i}{2}\sum_{m=j-1}^{j+1}\widetilde{\rho}_{m,k}\Omega_{j,m}e^{-i\varphi_{m-j}}-\frac{i}{2}\sum_{n=k-1}^{k+1}\rho_{j,n}\Omega_{k,n}e^{i\varphi_{n-k}}+\Gamma\sum_{l,p=j-1,k-1}^{j+1,k+1}\sqrt{\sigma_{j,l}\sigma_{k,p}}\rho^e_{l,p}, \tag{2}$$

$$\dot{\rho}_{j,k}=\frac{i}{2}\sum_{m=j-1}^{j+1}\rho^e_{m,k}\Omega_{j,m}e^{-i\varphi_{m-j}}-\frac{i}{2}\sum_{n=k-1}^{k+1}\rho^g_{j,n}\Omega_{n,k}e^{-i\varphi_{k-n}}-\Gamma\rho_{j,k}/2, \tag{3}$$

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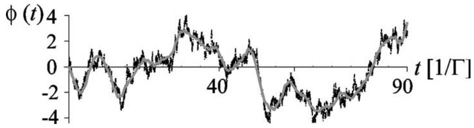

FIG. 1. A phase function obtained by sinusoidal fitting of the results of application of Eq. (6) (gray curve). The parameter values are $\Delta t = 0.01/\Gamma$, $1/B = 2/\Gamma$, and $1/\beta = 10/\Gamma$.

$$\dot{\tilde{\rho}}_{j,k} = \frac{i}{2}\sum_{m=j-1}^{j+1} \rho^{g}_{k,m}\Omega_{m,j}e^{i\varphi_{j-m}} - \frac{i}{2}\sum_{n=k-1}^{k+1} \rho^{e}_{n,j}\Omega_{k,n}e^{i\varphi_{n-k}} - \Gamma\tilde{\rho}_{j,k}/2, \tag{4}$$

where $\tilde{\rho}_{j,k} = \rho^{*}_{k,j}$, and Γ stands for the common spontaneous decay rate. The spontaneous-emission terms follow as usual [4,11]. The Rabi frequencies are defined as

$$\Omega_{k,j} = \sqrt{\sigma_{k,j}}\Gamma\mathcal{E}_{j-k}(\vec{r})/\sqrt{2I_s}, \tag{5}$$

where $I_s \equiv 2\pi^2\hbar c\Gamma/3\lambda^3$ is the saturation intensity and the parameter $\sigma_{k,j}$ denotes the branching ratio of the atomic transitions from state j to state k. The amplitudes $\mathcal{E}_{j-k}(\vec{r})$ are taken to be constants, and the Rabi frequencies real and nonnegative [1]. In Eqs. (1)–(4), those Rabi frequencies and branching ratios are zero in which the first subindex is not in the range of m_F's or the second subindex is not in the range of $\tilde{m}_F$'s. If the behavior of the complex amplitudes $\mathcal{E}_j(\vec{r})e^{i\varphi_j(\vec{r},t)}$ of the field components is known or properly modeled, one can solve Eqs. (1)–(4) and find the evolution of any of the magnetic-state populations.

In the following we apply the model to several examples. Let the field be composed of three phase-fluctuating laser beams which behave in the interaction region as plane waves, i.e., the field amplitude for each of the beams can be written as $E_j = \bar{\mathcal{E}}_j e^{-i\vec{k}_j\cdot\vec{r}+i\phi_j(t)}$, where $\vec{k}_j$ is the wave vector and $\phi_j(t)$ the fluctuating phase. Each of the beams is assumed to originate from a single-mode laser with a constant amplitude and the phase evolving in accordance with the Ornstein-Uhlenbeck phase-diffusion model [7–10,12–14]. We take no net drift for the phase, so that the light is on resonance between the two energy levels except for the frequency fluctuations originating from the phase fluctuations. Specifically, starting with a given phase at time t, $\phi(t)$, the phase at $t+\Delta t$ is to the first order given by [12–14]

$$\phi(t+\Delta t) = \phi(t)e^{-\beta\Delta t} + g\sqrt{b(1-e^{-2\beta\Delta t})/2\beta}. \tag{6}$$

The parameter $1/\beta$ characterizes the mean time of phase deviations and b conveys the bandwidth of the frequency fluctuations; with the choice $b = (B+\sqrt{B^2+16\beta B\ln 2})^2/(32\beta\ln 2)$ the bandwidth equals B [8,12]. Finally, g is a random number from a normal distribution with zero mean and variance equal to 1. In the calculations we set the time step to $\Delta t = 0.01/\Gamma$, while the parameters $1/\beta$ and $1/B$ have values greater than $1/\Gamma$. Each step is associated with an abrupt jump of ϕ. Solving Eqs. (1)–(4) numerically is substantially expedited by making the phase a smooth function of t. For this, we fitted the calculated data with a set of sinusoidal functions, excluding high frequencies, as shown in Fig. 1; small and fast deviations of ϕ, like those caused by the finite-step discretization in Eq. (6), have a negligible influence on the populations. The correspondence between the average absolute value of the derivative $\partial\phi(t)/\partial t$ and the field bandwidth B was carefully checked in each particular calculation.

As an example of application of the model we consider a four-state, two-level system with a single excited and three ground magnetic states, $F=1 \rightarrow \tilde{F}=0$. This is the simplest possible system that can respond to all three polarization components of light, and at the same time supports dark states. In fact, in this system, there is one-to-one correspondence between the components of the field and the magnetic states of the lower level. For instance, the E_{+1} circular component couples only to the $m_F=-1$ state. Moreover, this system can be realized in practice, e.g., by subjecting atoms of ^{87}Rb in the lower hyperfine ground state $|5\,^2\mathrm{S}_{1/2}, F=1\rangle$ to on-resonance radiation that drives atomic transitions to the state $|5\,^2\mathrm{P}_{3/2}, \tilde{F}=0\rangle$. Equations (1)–(4) in this case contain only summations over the ground magnetic states. The branching ratios $\sigma_{j,0}$ are all equal to 1/3. Equation (1), e.g., takes the form

$$\dot{\rho}^{e}_{0,0} = \frac{i}{2}\sum_{k=-1}^{+1} \Omega_{k,0}(\rho_{k,0}e^{i\varphi_{-k}} - \tilde{\rho}_{0,k}e^{-i\varphi_{-k}}) - \Gamma\rho^{e}_{0,0}, \tag{7}$$

and Eq. (2) with $k=j$ is

$$\dot{\rho}^{g}_{j,j} = \frac{i}{2}\Omega_{j,0}(\tilde{\rho}_{0,j}e^{-i\varphi_{-j}} - \rho_{j,0}e^{i\varphi_{-j}}) + \frac{1}{3}\Gamma\rho^{e}_{0,0}, \tag{8}$$

where j runs from -1 to $+1$. Initially the ground states are assumed to be equally populated. Then, the atoms are illuminated with laser beams which have orthogonal polarizations along either the Cartesian or spherical basis vectors. For each beam j, the phase is written as $\varphi_j(\vec{r},t) = -\vec{k}_j\cdot\vec{r} + \phi_j(t)$, where we assume that the fluctuating term $\phi(t)$ is common for all three beams, as it would be if they were derived from a single laser. Then we allow for certain time delays T_j between the beams such that $\phi_j(t) = \phi(t+s_jT_j)$, where s_j is equal to 0 for one beam and ± 1 for the other two beams. This scheme allows setting an arbitrary mutual correlation between the orthogonal vector components of the field. At $T_{\pm 1}=0$ the fluctuations of the field components are fully synchronized, and at large enough $T_{\pm 1}$ the fluctuations become independent.

Let first only two laser beams with their electric-field vectors directed along any two of the spherical basis vectors illuminate the atoms. This case has been considered in terms of incoherent depopulation pumping, e.g., in [15]. The magnetic state whose dipole moment of excitation is parallel to the third basis vector is not coupled to the field and, therefore, it will be permanently dark. Since the branching ratios $\sigma_{j,0}$ are equal, Eqs. (1)–(4) are symmetric with respect to the ground magnetic states $|\psi_{-1}\rangle$, $|\psi_0\rangle$, and $|\psi_{+1}\rangle$. Therefore, no matter along which two of the spherical basis vectors the beams are polarized, we can define the stable dark state as $|\psi_0\rangle$. Figure 2(a) shows the dynamics of the populations $\rho^{g}_{0,0}$ and $\rho^{e}_{0,0}$ of this dark state and the excited state, respectively, when the field bandwidth B is equal to $\Gamma/2$, $\beta=\Gamma/10$, and

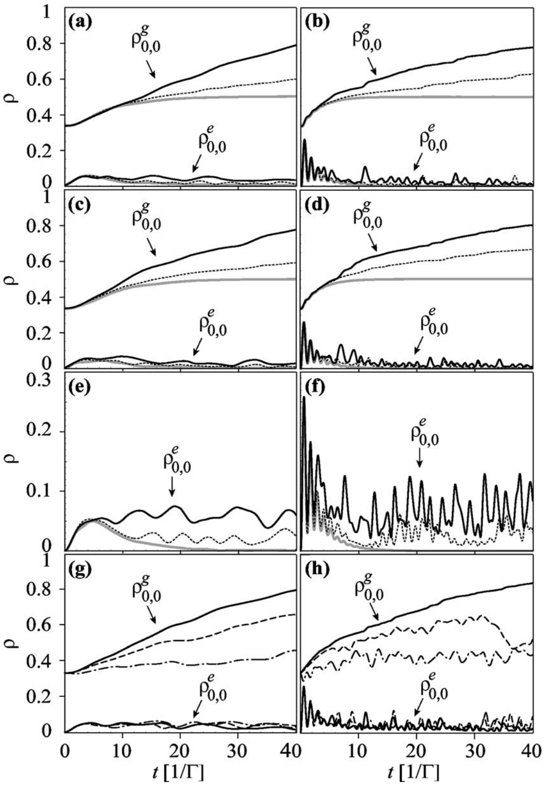

FIG. 2. Population dynamics in a four-state atom system. The total intensity of the beams, I_{tot}, is equal to $0.3\mathcal{A}$ for the figures on the left and $30\mathcal{A}$ on the right. $\rho^g_{0,0}$ and $\rho^e_{0,0}$ denote the populations of a stable dark state and the excited state, respectively. The degree of polarization of the field is 1 (gray curves), 0.8 (dotted curves), and 0.1 (solid black curves). The vector components of the field are directed in (a), (b), (e)–(h) along the spherical basis vectors and in (c) and (d) along Cartesian axes. The field has two orthogonal components in (a)–(d) and three components in (e)–(h). In (g) and (h), the intensity of one of the components is 0 (solid lines), $0.05I_{tot}$ (dashed lines), and $0.2I_{tot}$ (dash-dotted lines).

both intensities $I_{\pm 1} \equiv \bar{\mathcal{E}}^2_{\pm 1} = \mathcal{A}(\Omega_{\mp 1,0}/\Gamma)^2$ are equal to $0.15\mathcal{A}$ [see Eq. (5) with $2I_s/\sigma_{\mp 1,0} \equiv \mathcal{A}$]. For comparison, the saturation intensity $I_s = \sigma_{\mp 1,0}\mathcal{A}/2$ is equal to $0.17\mathcal{A}$. The figure illustrates three cases marked with gray, dotted, and solid black lines for which the two-dimensional (2D) degree of polarization [16] of the total field is $P_{2D}=1, 0.8$, and 0.1. Here, the degree of polarization P_{2D} is equal to the absolute value of the normalized correlation function of the beams, $|\langle e^{-i\{\phi(t)-\phi(t-T)\}}\rangle|$. The angular brackets denote time averaging, which was performed numerically with t running from 0 to $40/\Gamma$. The delay time T was adjusted until the degree of polarization attained a predetermined value. Given the degree of polarization, the corresponding pair of curves in Figs. 2(a)–2(h) was then obtained from a single run of the program. While the behavior of $\phi(t)$ is different in each run, the averaging over many runs essentially only washes out the fast-term fluctuations of the populations.

If the beams are fully correlated, the atoms are pumped into two dark states [4], of which one is $|\psi_0\rangle$ and the other a superposition of $|\psi_{-1}\rangle$ and $|\psi_{+1}\rangle$. Hence, the population $\rho^g_{0,0}$, shown by the upper gray curve of Fig. 2(a), saturates to the value of 0.5, and the excited-state population $\rho^e_{0,0}$ (the lower gray curve) decays to zero within a time of several $1/\Gamma$. If the beams are not fully correlated, the excited-state population shows an oscillating behavior, due to the polarization fluctuations, and gradually all the atoms are pumped into the single state $|\psi_0\rangle$. A small decrease of the mutual correlation of the beams destroys the competing dark state. When P_{2D} decreases further, the pumping rate to $|\psi_0\rangle$ increases. This rate, on the other hand, is not sensitive to the bandwidth B at a fixed P_{2D}. Since in this particular level scheme one can always transform away any constant phases in the interaction matrix elements by redefining the phases of the state vectors spanning the ground level, the phases $-\vec{k}_j \cdot \vec{r}$ do not influence the population dynamics. The pumping rate will therefore be spatially uniform even if the field is created by two laser beams which propagate in perpendicular directions.

Next, we increase the intensities $I_{\pm 1}$ by two orders of magnitude and obtain the curves in Fig. 2(b). Within a short time $t < 10/\Gamma$, the population of $|\psi_0\rangle$ rises quickly, but then it evolves essentially in the same manner as in the previous example. This insensitivity of the pumping rate to the field intensity is partially explained by the constant rate of spontaneous emission. On the other hand, at each time moment the atoms are pumped not only into $|\psi_0\rangle$ but also into a superposition dark state in which, independently of the intensity, the atoms stay decoupled from the field until the polarization changes. Then the dark state is destroyed, and the stronger the field is the more rapid can be the increase of $\rho^e_{0,0}$. However, simultaneously, depopulation pumping to the new dark state takes place. As a result, $\rho^e_{0,0}$ shows spikes, but the continuous increase of $\rho^g_{0,0}$ remains similar to that taking place at lower intensity levels and equal P_{2D}.

Another example is illustrated in Figs. 2(c) and 2(d). This time the beams are polarized along the two Cartesian basis vectors $\vec{x}$ and $\vec{y}$. The quantization axis is the z axis, and the permanently dark state $|\psi_0\rangle$ is the state with $m_F=0$. In the previous example, this state was pumped with atoms by the action of two equal-amplitude phase-fluctuating waves with opposite circular polarizations. Therefore, for narrowband fluctuations, the total field could be considered as linearly polarized with a randomly oscillating plane of polarization. In the present case, the total field can have an arbitrary elliptical polarization, and the phase fluctuations lead to random changes of the ellipticity. Setting the field amplitudes $\bar{\mathcal{E}}_x$ and $\bar{\mathcal{E}}_y$ to the same values as the amplitudes $\bar{\mathcal{E}}_j$ in the previous calculations and solving Eqs. (1)–(4), with $\mathcal{E}_{\pm 1}$ being written in terms of $\bar{\mathcal{E}}_x$ and $\bar{\mathcal{E}}_y$, we obtain the curves in Figs. 2(c) and 2(d). The behavior of $\rho^g_{0,0}$ is practically indistinguishable from that illustrated in Figs. 2(a) and 2(b), including the insensitivity of the results to the propagation-gained phases and to the bandwidth. Thus, in an on-resonance two-vector-component field, the long-term dynamics of the dark ground state and of the excited state, e.g., the time scale over which the populations approach their final values, is mainly governed by the degree of polarization rather than by the mean polarization state, the bandwidth, or the intensity of the light.

We proceed by adding a third vector component to the field. Let the three beams be polarized along the spherical basis vectors (linear polarizations yield similar results) and their intensities be equal to $0.1\mathcal{A}$ each. If the T_j's are zero, the fields are fully correlated. The resulting two dark states are pumped with atoms and the population of the excited state drops to zero [see the gray line in Fig. 2(e)]. The dotted and black solid lines show what happens if the 3D degree of polarization of the total field decreases to 0.8 and 0.1, respectively. The degree of polarization is calculated from $P_{3D}=\sqrt{(p_{12}^2+p_{23}^2+p_{13}^2)/3}$, where p_{ij} denotes the degree of correlation of the beams i and j, and, therefore, p_{ij} is equivalent to the two-dimensional degree of polarization P_{2D} [16]. As seen in Fig. 2(e), the excited-state population of the atoms oscillates around a level determined by P_{3D}. A similar behavior of $\rho_{0,0}^e$ is obtained for atoms interacting with a field of 100 times higher intensity [see Fig. 2(f)]. The strong spiking of $\rho_{0,0}^e$ is caused by the high values of the Rabi frequencies. Such Rabi oscillations of the populations reflect an interplay between the field intensity and polarization fluctuations. The oscillations are not damped out by spontaneous emission, as they would if the field was fully polarized.

As an intermediate case between the two cases already discussed, we finally show how an additional weak orthogonal field component can destroy the population trapping by a 2D field. The black solid curves in Figs. 2(g) and 2(h) are calculated under the same conditions as the ones in Figs. 2(a) and 2(b). We then move 5% and, afterward, 20% of the field intensity into the third orthogonal component. The results are shown by the dashed and dash-dotted lines, respectively. Obviously the state $|\psi_0\rangle$ can no longer be considered as a stable dark state. The magnetic-state populations remain close to their initial values if the intensity of the third component exceeds 20% of the total field intensity.

For a fully polarized radiation, our system gets pumped into two dark states at a time scale that depends on light intensity and spontaneous emission rate. When the polarization fluctuates slowly (narrow band) in a plane, one of the dark states changes with the fluctuations and releases its population. Eventually the system gets pumped into the remaining dark state at a rate that depends mainly on the degree of polarization of light. If the polarization fluctuates in all directions, there is no permanent dark state. Once more, after the initial transient, the population dynamics is governed by the fluctuations, here mainly by the three-dimensional degree of polarization. From a more formal point of view that could also serve as a starting point for mathematical analysis, there are three distinct time scales in the problem: infinity for the dark states, inverse linewidth of the radiation, and the usual time scales of a two-level system. No matter how narrowband the field is, polarization fluctuations remove the infinite time scale. After an initial transient, populations then evolve governed by the fluctuations.

In general terms, we have studied light-driven transitions between energy levels that exhibit angular momentum degeneracy. We have shown that in the presence of dark states even narrowband phase fluctuations may dramatically influence the dynamics of magnetic-state populations; one of the quantities characterizing this influence is the degree of polarization of the field. The knowledge of the interaction features described in this work can be useful for experimentalists dealing with on-resonance optical pumping.

We acknowledge financial support from the Academy of Finland, NSF, and NASA.

[1] B. W. Shore, Phys. Rev. A **17**, 1739 (1978).
[2] V. Milner, B. M. Chernobrod, and Y. Prior, Europhys. Lett. **34**, 557 (1996).
[3] V. Milner, B. M. Chernobrod, and Y. Prior, Phys. Rev. A **60**, 1293 (1999).
[4] A. V. Taichenachev *et al.*, Phys. Rev. A **69**, 033410 (2004).
[5] E. Arimondo, Prog. Opt. **35**, 257 (1996).
[6] P. Tremblay and C. Jacques, Phys. Rev. A **41**, 4989 (1990).
[7] K. Arnett *et al.*, Phys. Rev. A **41**, 2580 (1990).
[8] S. N. Dixit, P. Zoller, and P. Lambropoulos, Phys. Rev. A **21**, 1289 (1980).
[9] Th. Haslwanter *et al.*, Phys. Rev. A **38**, 5652 (1988).
[10] M. H. Anderson *et al.*, Phys. Rev. Lett. **64**, 1346 (1990).
[11] C. Cohen-Tannoudji, *Atoms in Electromagnetic Fields* (World Scientific Publishing, Singapore, 1994), Sec. 3.1.
[12] M. Høyrup, M.Sc. thesis, Aarhus University, 1996 (unpublished).
[13] D. T. Gillespie, Phys. Rev. E **54**, 2084 (1996).
[14] L. Bartosch, Int. J. Mod. Phys. C **12**, 851 (2001).
[15] W. Happer, Rev. Mod. Phys. **44**, 169 (1972).
[16] T. Setälä *et al.*, Phys. Rev. E **66**, 016615 (2002).