

Effects of Zeeman degeneracy on the steady-state properties of an atom interacting with a near-resonant laser field: Analytic results

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The analytic solution of the steady-state density matrix is presented for a closed two-level atom with an arbitrary ground-state angular momentum J_g and an excited-state angular momentum J_e interacting with a linearly polarized laser field. From this solution, analytic formulas for the total rate of laser-atom scattering, the rate of coherent scattering, and radiative forces are derived. Interestingly, it is found that population inversion in true atomic states, as opposed to the population inversion in the dressed states, can occur under certain conditions. A method is proposed to take advantage of this inversion for light amplification. The mathematical formulation is built around the concept of the resolvent operator in Liouville space, with some conclusions that are generally applicable to any system with a unique steady state.

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I. INTRODUCTION

Experimental and theoretical progresses in laser cooling and nonlinear spectroscopy have demonstrated clearly the important role of atomic Zeeman structure in near-resonant laser-atom interactions. It is, for example, responsible for polarization-gradient cooling [1–3] and for narrow features in weak-probe absorption spectra [4,5]. Theory for such experiments, assuming a closed two-level system, requires the solution of a density-matrix equation of dimension $[2(J_e + J_g + 1)]^2$, which is equal to 400 for the cycling transition of cesium with $J_g = 4$ and $J_e = 5$ (J stands for F here). Because we are primarily interested in the nonperturbative regime, the traditionally powerful method of irreducible representation [6,7] loses much of its edge since different K components (tensors of different rank) can be strongly coupled by the laser field. The dressed-state approach can simplify the problem a great deal [8] if only the Rabi splitting is much greater than the spontaneous decay width, which is however not the case in most experiments using laser traps. As a result, solutions which are valid for all laser intensities (limited only by the assumption of a *closed* two-level system) has been limited to a few specific cases [9,10]. In this paper, the analytic solution of the steady-state density matrix for a closed two-level atom with arbitrary ground-state angular momentum J_g and excited-state angular momentum J_e interacting with a linearly polarized laser field will be presented. The case of circular polarization is simple and will be discussed only briefly. Atomic motion will be ignored except for the trivial case of a plane wave. Elliptical polarization will not be considered. Just to be focused, only results which can be derived analytically are presented here. Anything requiring numerical computation, such as an emission or weak-probe-absorption spectrum, will be presented in a separate publication. Experimentalists who are not interested in the details of the derivation can go directly to

Eqs. (29)–(31) and subsequent discussions. For theorists, Sec. II should not be overlooked.

II. GENERAL RELATION BETWEEN STEADY-STATE DENSITY MATRIX AND RESOLVENT OPERATOR

Before we get to the specific problem of laser-atom interaction, let us first explore the relationship between the steady-state density matrix and the resolvent operator in a broader context. Consider a system described by the following density-matrix equation:

$$\partial_t \rho(t) = L \rho(t), \quad (1)$$

where L is some arbitrary time-independent Liouville operator. All we will assume about this system is that it is closed and supports a *unique* steady state ρ^{SS} . The resolvent operator (in the Liouville space) for this equation is defined as

$$R(s) \equiv [\mathcal{L}(s)]^{-1}, \quad (2)$$

where $\mathcal{L}(s) \equiv s - L$, and s is a complex variable. Solving Eq. (1) gives

$$\rho^{\text{SS}} = \lim_{t \rightarrow \infty} e^{Lt} \rho_0 = \lim_{s \rightarrow 0} s R(s) \rho_0, \quad (3)$$

with ρ_0 being some arbitrary initial density matrix. More explicitly, we have

$$\langle i | \rho^{\text{SS}} | j \rangle = \lim_{s \rightarrow 0} s \sum_{k,l} \langle \langle i, j | R(s) | k, l \rangle \rangle \langle k | \rho_0 | l \rangle. \quad (4)$$

This equation is, however, not very useful. It has ρ_0 on the right-hand side when in fact ρ^{SS} should have nothing to do with it. To get the real meaning behind this equation, it is important to remember that ρ_0 is completely arbitrary except that it is Hermitian and has its trace equal to 1 (i.e., $\sum_k \langle k | \rho_0 | k \rangle = 1$). It can then be shown that Eq. (4) actually implies

$$\lim_{s \rightarrow 0} s \langle \langle i, j | R(s) | k, l \rangle \rangle = 0, \quad (5)$$

for $k \neq l$ and arbitrary i, j , and

$$\langle i | \rho^{\text{SS}} | j \rangle = \lim_{s \rightarrow 0} s \langle i, j | R(s) | k, k \rangle, \quad (6)$$

for arbitrary i, j , and k . These two equations characterize the behavior of $R(s)$ near the point $s=0$. They specify which matrix elements of the resolvent operator have $1/s$ poles and relate the residues of those poles directly to the steady-state density matrix. These properties are true for any closed system [defined by Eq. (1)] with a unique steady state. Before using Eqs. (5) and (6) for our specific purposes in this paper, I will simply mention that they will also be very useful in implementing a steady-state perturbation theory which is needed for treating, for example, atom-atom interactions in a strong laser field [11]. The importance of the resolvent operator lies of course not only in its relationship with the steady-state density matrix as given by Eq. (6), but also in the fact that it is the quantity that we have to deal with when calculating any correlation functions using the quantum regression theorem [12,13].

$$\begin{aligned} \partial_t \rho_A^R(t) = L_A^R \rho_A^R(t) = -i & \left\{ \left[-(\Delta + i\gamma/2) \sum_{m_e} |J_e m_e\rangle \langle J_e m_e| \right] \rho_A^R(t) - \rho_A^R(t) \left[-(\Delta + i\gamma/2) \sum_{m_e} |J_e m_e\rangle \langle J_e m_e| \right]^\dagger \right\} \\ & + \gamma \sum_{q, \text{all } m} \langle J_g m'_g 1q | J_e m'_e \rangle |J_g m'_g\rangle \langle J_e m'_e | \rho_A^R(t) | J_e m_e \rangle \langle J_g m_g | \langle J_g m_g 1q | J_e m_e \rangle \\ & - i \left[-(\Omega_{eg}/2) \sum_m f_m (|J_e m\rangle \langle J_g m| + |J_g m\rangle \langle J_e m|), \rho_A^R(t) \right], \end{aligned} \quad (7)$$

where $\Delta = \omega - \omega_{eg}$ is the laser detuning; γ is the spontaneous decay rate of the excited state; $\Omega_{eg} = E_0 \langle J_e || \mu || J_g \rangle / \hbar$ is the reduced Rabi frequency; and

$$f_m \equiv (-1)^{J_e - m} \begin{bmatrix} J_e & 1 & J_g \\ -m & 0 & m \end{bmatrix}.$$

Note that J is merely a notation which can also stand for F (the total angular momentum including nuclear spin). It is easy to show that the representation of L_A^R [and therefore $\mathcal{L}(s)$ and $R(s)$] in the basis of $|J' m', J m\rangle$ is block diagonal in $l \equiv (m' - m)$. This is of course what one would expect because of the cylindrical symmetry of the problem. Using Eq. (5), we see immediately that there will be no coherences between states having different m values (this is only true in the coordinate system that we have chosen). All the information about the steady-state density matrix is then contained solely in the $l=0$ manifold of L_A^R (for emission spectra and absorption spectra by a weak probe, the $l=\pm 1$ manifolds will also come into play). There are $4J_g + 1$ l manifolds (because for $J_e = J_g + 1$, it can be shown that states having to do with either $|J_e J_e\rangle$ or $|J_e - J_e\rangle$ are basically irrelevant), each having a dimension of $4(2J_g + 1 - |l|)$. Letting

III. ANALYTIC SOLUTION FOR NEAR-RESONANT LASER-ATOM INTERACTION

We now focus on the specific problem of a stationary two-level atom with Zeeman degeneracy interacting with either a circularly or a linearly polarized coherent laser field. The case of circular polarization is rather "boring", since the atom will either be pumped into a dark state (if $J_e \leq J_g$) or become a simple two-level system (if $J_e > J_g$) which has been well studied [13–15]. We will simply ignore it here (noting that either collision [10] or a magnetic field can make it interesting). For linear polarization, the case where $J_e < J_g$ is again straightforward. The atom simply gets pumped into dark states $|J_g - J_g\rangle$ and $|J_g + J_g\rangle$ with equal population. The rest of this paper will therefore deal only with the case of linear polarization and $J_e \geq J_g$.

For a stationary atom in a coherent laser field linearly polarized in the z direction, the density-matrix equation in the rotating frame (in which L_A^R is time independent) is given by [16]

$\mathcal{L}^{(l)}(s)$ be the representation of $\mathcal{L}(s) [\equiv s - L_A^R]$ in each l manifold, it has the following form for all l :

$$\mathcal{L}^{(l)}(s) = \begin{bmatrix} aI & A^{(l)} & B^{(l)} & C^{(l)} \\ A^{(l)} & bI & 0 & B^{(l)} \\ B^{(l)} & 0 & cI & A^{(l)} \\ 0 & B^{(l)} & A^{(l)} & dI \end{bmatrix}. \quad (8)$$

Here a, b, c, d are defined by $a = s$, $b = s - i\Delta + \gamma/2$, $c = s + i\Delta + \gamma/2$, and $d = s + \gamma$. I represents the unit matrix. $A^{(l)}$, $B^{(l)}$, and $C^{(l)}$ are all matrices of dimension $(2J_g + 1 - |l|)$ with their elements defined by

$$\langle m' | A^{(l)} | m \rangle \equiv - \langle \langle J_e m', J_g m' - l | L_A^R | J_g m, J_g m - l \rangle \rangle, \quad (9)$$

$$\langle m' | B^{(l)} | m \rangle \equiv - \langle \langle J_g m', J_e m' - l | L_A^R | J_g m, J_g m - l \rangle \rangle, \quad (10)$$

$$\langle m' | C^{(l)} | m \rangle \equiv - \langle \langle J_g m', J_g m' - l | L_A^R | J_e m, J_e m - l \rangle \rangle. \quad (11)$$

It should be quite obvious that both $A^{(l)}$ and $B^{(l)}$ are diagonal matrices, and $C^{(l)}$ is tridiagonal. The corresponding $R^{(l)}(s)$ can be written as

transition, our solution reduces of course to that of an idealized two-level atom, with $w_0=1$, $f_0=1/\sqrt{3}$, and $g_s=\frac{1}{3}$.

The following are some more of the characteristics of Eqs. (29)–(31). First,

$$\langle J_g - m | \rho_A^{SS,R} | J_g - m \rangle = \langle J_g m | \rho_A^{SS,R} | J_g m \rangle, \quad (32)$$

which is of course what one would expect for a cylindrically symmetric system. Second,

$$\langle J_e - m | \rho_A^{SS,R} | J_g - m \rangle = (-1)^{J_e - J_g + 1} \langle J_e m | \rho_A^{SS,R} | J_g m \rangle. \quad (33)$$

Third, the total upper-state population, given by

$$P_e(\Delta) = \sum_m \langle J_e m | \rho_A^{SS,R} | J_e m \rangle \\ = (g_s \Omega_{eg}^2 / 4) [\Delta^2 + (\gamma/2)^2 + g_s \Omega_{eg}^2 / 2]^{-1}, \quad (34)$$

is never greater than $\frac{1}{2}$. It approaches $\frac{1}{2}$ for large Ω_{eg} . The total upper-state population is an important quantity since it is directly related to both the total laser photon scattering (extinction) rate and the radiation pressure force (see next section). A nice feature of Eq. (34) is that all geometrical dependences are summarized in a single parameter g_s . Its values for all transitions of interest, up to $J_g=4$ and $J_e=5$, are given in Table I.

Note that in the limit of large detuning ($\Delta \gg \gamma, \Omega_{eg}$) or the limit of weak field ($\Omega_{eg} \rightarrow 0$), Eqs. (29)–(31) reduce to

$$\langle J_e m | \rho_A^{SS,R} | J_g m \rangle = 0, \quad (35)$$

$$\langle J_e m | \rho_A^{SS,R} | J_e m \rangle = 0, \quad (36)$$

$$\langle J_g m | \rho_A^{SS,R} | J_g m \rangle = w_m, \quad (37)$$

where w_m is not equal to $1/(2J_g+1)$ except for the cases of $J_g=0$ and $J_g=\frac{1}{2}$. This result may look surprising at first. But think about it. All it is saying is that no matter how weak the laser field is, given sufficient interaction time (which we have assumed to be infinite), the atom will eventually get pumped, which we resentfully accept. However, any discomfort we feel at this point is perfectly

legitimate. The point is that Eqs. (29)–(31) are valid only *theoretically* for the infinitely weak laser field (and large detuning), under the assumption of infinite interaction time. They also break down if Ω_{eg} is exactly zero because our basic assumption about the existence of a *unique* steady state would no longer be valid. In more practical terms, Eqs. (29)–(31) apply only when optical pumping dominates over other processes such as collisions and that the laser-atom interaction time is long enough to ensure that the steady state can be reached. These conditions are well satisfied in most laser traps, and can often be forced upon other situations.

IV. OTHER RESULTS

Here I present some other results which can be derived directly from our analytic solution. The derivations are relatively straightforward. I will therefore simply give the results. The total rate of photon scattering is directly related to the total upper-state population $P_e(\Delta)$ and is given by

$$W_t(\Delta) = \gamma P_e(\Delta) = \gamma (g_s \Omega_{eg}^2 / 4) \\ \times [\Delta^2 + (\gamma/2)^2 + g_s \Omega_{eg}^2 / 2]^{-1}. \quad (38)$$

The radiation pressure force on an atom moving with velocity \mathbf{v} by a linearly polarized plane wave is given by

$$\mathbf{F}(\Delta, \mathbf{v}) = \hbar \mathbf{k}_L W_t(\Delta - \mathbf{k}_L \cdot \mathbf{v}), \quad (39)$$

where \mathbf{k}_L is the k vector of the laser field. The rate of coherent scattering, which is related to steady-state ground- and excited-state coherences $\langle J_e m | \rho_A^{SS,R} | J_g m \rangle$, is given by

$$W_c(\Delta) = (2J_e + 1) \gamma (g_s^2 \Omega_{eg}^2 / 4) [\Delta^2 + (\gamma/2)^2] \\ \times [\Delta^2 + (\gamma/2)^2 + g_s \Omega_{eg}^2 / 2]^{-2}. \quad (40)$$

Note that in the limit of large detuning, the total rate of photon scattering W_t is *not* equal to the rate of coherent scattering except for the case of $J_g=0$ (ideal two-level system). This is because for all atoms with $J_g \neq 0$ we have the possibility of spontaneous Raman scattering (in addition to coherent Raleigh scattering) even in the limit of large detuning. Finally, the force on a stationary atom at position \mathbf{x} in a linearly polarized standing wave is given by

$$\mathbf{F}(\Delta) = 2\hbar \mathbf{k}_L \Delta \tan(\mathbf{k}_L \cdot \mathbf{x}) [g_s \Omega_{eg}^2(\mathbf{x}) / 4] \\ \times [\Delta^2 + (\gamma/2)^2 + g_s \Omega_{eg}^2(\mathbf{x}) / 2]^{-1}, \quad (41)$$

where $\Omega_{eg}(\mathbf{x}) \equiv \Omega_{eg}(0) \cos(\mathbf{k}_L \cdot \mathbf{x})$.

V. FURTHER DISCUSSIONS

For the properties discussed so far in this paper, an atom with Zeeman degeneracy bears a great deal of resemblance to an idealized two-level system. For example, the expressions for the forces have exactly the same form as the forces for a simple two-level atom with no Zeeman degeneracy. All geometrical dependences are contained in the parameter g_s , which simply scales the

TABLE I. Values for g_s .

J_g	J_e	g_s
0	1	0.333 333
$\frac{1}{2}$	$\frac{1}{2}$	0.166 667
$\frac{1}{2}$	$\frac{3}{2}$	0.166 667
1	2	0.117 647
$\frac{3}{2}$	$\frac{3}{2}$	0.03
$\frac{3}{2}$	$\frac{5}{2}$	0.093 333 3
2	3	0.078 091 1
$\frac{5}{2}$	$\frac{5}{2}$	0.012 410 4
$\frac{5}{2}$	$\frac{7}{2}$	0.067 346 9
3	4	0.059 268 5
$\frac{7}{2}$	$\frac{7}{2}$	0.006 774 54
$\frac{7}{2}$	$\frac{9}{2}$	0.052 943 4
4	5	0.047 847 4

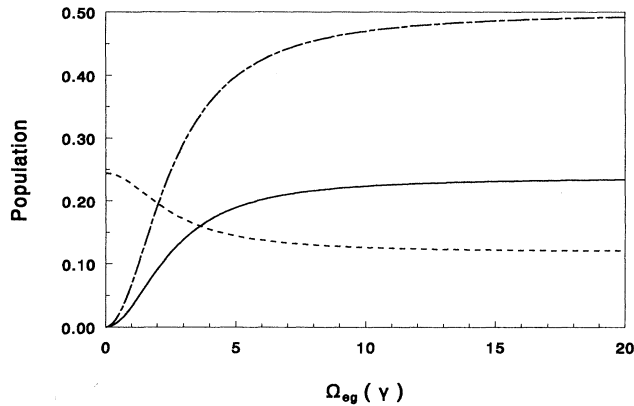


FIG. 1. Populations for the $J_g=2$ to $J_e=3$ transition at zero detuning. Solid line: population in the $|J_e, 0\rangle$ state; dashed line: population in the $|J_g, 1\rangle$ state; dash-dotted line: total population in the excited state.

Rabi frequency. This gives some explanation as to why the two-level-system model has been so successful in describing radiation pressure force on a real atom. However, this resemblance does not go very far from here. The first thing that comes to mind is obviously angular distribution of scattered radiation. It is not much of an issue for an ideal two-level system ($J_g=0$, $J_e=1$) mainly because it is frequency independent, which will however no longer be the case for any transition with $J_g \neq 0$. There are many other interesting differences. For absorption spectrum of a weak-probe beam, population differences created by the pump beam among different ground-state Zeeman sublevels give rise to narrow structures due to stimulated Raman scattering [4,5]. No counterpart of these structures exists in corresponding two-level-atom spectra [14,17]. Also, the radiative force by a standing wave will be quite different from the simple two-level-atom case when motion is to be considered.

Finally, Eqs. (29)–(31) present an interesting possibility for population inversion between states $|J_e m\rangle$ and $|J_g m+1\rangle$ (or $|J_g m-1\rangle$). This is *somewhat* surprising since the total population in the excited state is always less than the total population in the ground state. To be specific, let us look at the case of $J_g=2$ and $J_e=3$, relevant for the cycling transition of the sodium atom. The populations in $|J_e 0\rangle$ and $|J_g 1\rangle$ as well as the total population in the excited state at zero detuning are plot-

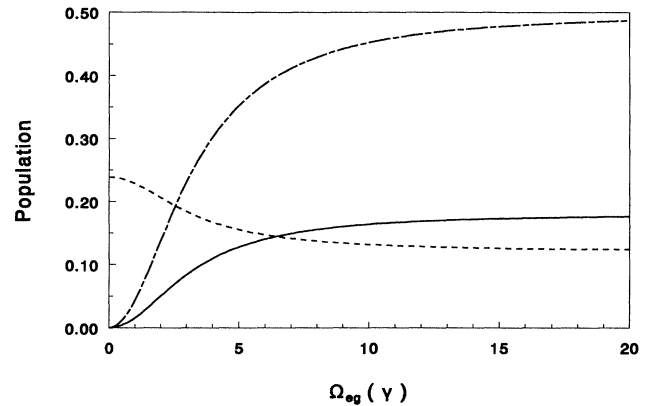


FIG. 2. Same as Fig. 1 except for $J_g=4$ and $J_e=5$.

ted in Fig. 1. One can see that as the Rabi frequency increases beyond some critical value, the population in $|J_e 0\rangle$ becomes greater than the population in $|J_g 1\rangle$. From Fig. 2 we see that the same happens for the cycling transition of cesium ($J_g=4$, $J_e=5$). The critical Rabi frequencies are achievable since they are still below the value for saturation. With the setup that we have been concerned with, this inversion does not yet lead to any consequences such as light amplification, because it is overshadowed by the population difference between $|J_e-1\rangle$ and $|J_g 0\rangle$. This problem can be easily fixed, however. For example, by adding a magnetic field in the direction of the laser polarization we can shift these two transitions out of resonance from each other. For such a medium, what one would observe at this frequency (with the presence of the magnetic field properly taken into account) is that as the pump-laser intensity increases, the medium turns from absorptive to amplifying for the circular polarization component of a probe beam. This amplification scheme, with its cross section roughly proportional to Ω_{eg}^2 , is expected to be more efficient at small pump intensity than that based on population inversion in the dressed states [14,17], which have cross sections roughly proportional to Ω_{eg}^4 .

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