Coherent irradiation of multilevel atoms in branched and cyclic configurations

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An analytic solution is presented for the time development of an n-level atom in resonant interaction with $n-1$ lasers in a branch configuration, which is an alternative generalization of the three-level atom to the commonly studied cascade configuration. The same analytic approach is also applied to a four-laser resonant interaction with a four-level atom in a cyclic configuration, where it is found that a change of phase of any one of the lasers has a critical effect on the population dynamics and, with certain Rabi frequency and laser phase relationships, double-quantum transitions can be eliminated completely.

I. INTRODUCTION

The interaction of atoms with coherent fields, such as those produced by single-mode lasers, has been the subject of intensive study, with much of the work devoted to two-level atoms. The interaction of three-level atoms[1–3] with two fields has also received much attention. Some attention has been given to four- and higher-level cases where the emphasis has been on cascade-type configurations,[4,5] though other configurations have also been studied.[6–8]

In this paper we examine some multilevel configurations for which the time evolution can be solved analytically, with the obvious advantage over numerically solved cases in giving an understanding of the processes involved.

The first configuration we examine is the branch configuration shown in Fig. 1, in which $n-1$ of the n levels are coupled by means of $n-1$ lasers to a common level. This can be seen as an alternative generalization of the three-level case to the cascade configuration. Later we examine another configuration, a cyclic configuration, where the phases of the laser fields become important. We consider only timescales short enough for decay to be neglected and assume that the Rabi frequencies are large enough for a few population cycles to occur in such timescales. We also assume that the levels are sufficiently unevenly spaced, compared with the power broadening, for each laser to drive only one transition, which is driven on resonance, and any nonresonant level shifts are either negligible or already incorporated into the level spacings. Our approach is to solve for the time-displacement operator $U(t,t_0)$. Once this is found the evolution of the density matrix or any Heisenberg operator can be found; however, here we shall limit ourselves to finding transition probabilities.

II. METHOD OF SOLUTION

The time-displacement operator is found by the method of transformations,[9] in which an unitary transformation is made by means of an operator T to a reference frame in which the effective Hamiltonian $\mathscr{H}$ is static. The time-displacement operator in this frame is then, in units with $\hbar=1$,

$$\bar{U}(t,t_0)=\exp[-i\mathscr{H}(t-t_0)] , \quad (1)$$

and in the laboratory frame it is

$$U(t,t_0)=T^{-1}(t)\bar{U}(t,t_0)T(t_0) . \quad (2)$$

The formal solution (1) involves an infinite series of terms in powers of $\mathscr{H}$ and as such is not generally immediately applicable to expressing transition probabilities exactly in a finite number of terms. If various recurrence relations exist, however, the infinite series (1) can be replaced by a finite series. The simplest such relation is

$$\mathscr{H}^2=k^2I , \quad (3)$$

where I is the unit operator and k is a constant, with which (1) becomes

$$\bar{U}(t,t_0)=\cos[k(t-t_0)]-i\mathscr{H}k^{-1}\sin[k(t-t_0)] . \quad (4)$$

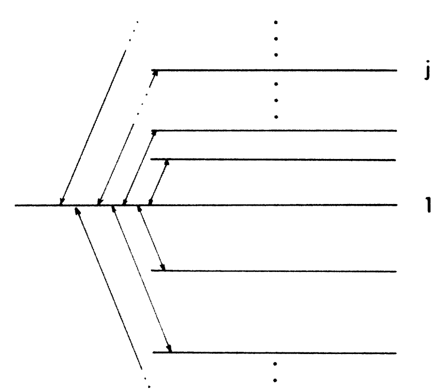

FIG. 1. Energy-level diagram of an n-level system in resonant interaction with $n-1$ lasers in a branch configuration. The arbitrary zero level of energy is set at level 1, which is the common level coupled to all other levels. The laser associated with Rabi frequency α_j couples levels 1 and j.

An example of this is where $\mathscr{H}$ is proportional to an effective spin-$\frac{1}{2}$ operator, as for a two-level system.

Another simple recurrence relation is

$$\mathscr{H}^3 = b^2 \mathscr{H} , \tag{5}$$

for which the series (1) becomes[2]

$$\bar{U}(t,t_0) = 1 + b^{-2}\mathscr{H}^2\{\cos[b(t-t_0)] - 1\} - ib^{-1}\mathscr{H}\sin[b(t-t_0)] . \tag{6}$$

This can occur, for example, if $\mathscr{H}$ is proportional to a spin-1 operator.

The multilevel configurations studied in this paper are such that the static effective Hamiltonian obeys either recurrence relation (3) or recurrence relation (5).

III. BRANCH CONFIGURATION

With the levels coupled as shown in Fig. 1, we choose level 1 to be the common level coupled to all other levels, and set our arbitrary zero of energy at this level. The laboratory Hamiltonian can thus be written in the general form

$$\mathscr{H}_L = \sum_{j\,(\neq 1)} [\,|j\rangle\langle j\,|E_j + 2\alpha_j \cos(\omega_j t + \phi_j) \times (\,|1\rangle\langle j\,| + \text{H.c.})] , \tag{7}$$

where α_j is the resonant Rabi frequency for the coupling of level 1 with level j, and ω_j and ϕ_j are the frequency and phase of the laser beam j. E_j is the energy of level j. Without losing generality, we have made α_j in (7) real and positive, which can be done by choosing the appropriate quantum-mechanical phase factor $\exp(i\beta_j)$ of state $|j\rangle$ to combine with any phase factor associated with α_j to give unity. The associated unitary transformation is

$$\exp\left[i \sum_{j\,(\neq 1)} |j\rangle\langle j\,|\beta_j\right] .$$

The unitary transformation we use to obtain a static Hamiltonian is

$$T = \exp\left[i \sum_{j\,(\neq 1)} |j\rangle\langle j\,|(\omega_j t + \phi_j)\right] , \tag{8}$$

with the transformed Hamiltonian then given by

$$\mathscr{H} = T\mathscr{H}_L T^{-1} + i\dot{T}T^{-1} . \tag{9}$$

Substituting (8) into (9) and discarding terms oscillating at twice the optical frequencies, which produce optical Bloch-Siegert effects, we find that $\mathscr{H}$ simplifies, with the resonance condition $E_j = \omega_j$, to the static form

$$\mathscr{H} = \sum_{j\,(\neq 1)} \alpha_j(\,|1\rangle\langle j\,| + |j\rangle\langle 1\,|\,) . \tag{10}$$

From this we obtain

$$\mathscr{H}^2 = \sum_{j\,(\neq 1)} \alpha_j \left[\alpha_j\,|1\rangle\langle 1\,| + \sum_{k\,(\neq 1)} \alpha_k\,|j\rangle\langle k\,|\right] \tag{11}$$

and, hence,

$$\mathscr{H}^3 = b^2 \mathscr{H} , \tag{12}$$

where

$$b^2 = \sum_{j\,(\neq 1)} \alpha_j^2 . \tag{13}$$

Thus we have a recurrence relation of the form (5), and hence a time-displacement operator of the form (6). The transformation (8) only alters the phases of the states, so from (2) we can find the transition probabilities directly from $\bar{U}(t,t_0)$, e.g., if the atom is initially in state $|1\rangle$ at time t_0, the probability P_{k1} that it is found in another state $|k\rangle$ at time t is

$$P_{k1} = |\langle k\,|\bar{U}(t,t_0)\,|1\rangle|^2 . \tag{14}$$

From (11), $\langle k\,|\mathscr{H}^2|\,1\rangle$ is zero, so from (6) and (10) we find that

$$P_{k1} = b^{-2}\alpha_k^2 \sin^2[b(t-t_0)] . \tag{15}$$

Similarly, we find the transition probabilities P_{kj} between states with $k \neq 1$, $j \neq 1$, and P_{11} to be

$$P_{kj} = b^{-4}\alpha_k^2\alpha_j^2\{\cos[b(t-t_0)] - 1\}^2 \tag{16}$$

and

$$P_{11} = \cos^2[b(t-t_0)] . \tag{17}$$

The modulus of the amplitude that the atom remains in an initial level $i \neq 1$ is

$$|\bar{U}_{ii}(t,t_0)| = 1 + \alpha_i^2 b^{-2}\{\cos[b(t-t_0)] - 1\} , \tag{18}$$

which has a minimum value of $1 - 2\alpha_i^2 b^{-2}$. Clearly, state $|i\rangle$ will never transfer its total population to the other states if the Rabi frequency α_i is sufficiently weak for $\alpha_i^2 < b^2/2$. This is a generalization of the effect described by Shore and Ackerhalt[3] for a three-level atom. When α_i is small, the corresponding laser beam can be considered as a weak probe beam probing level 1. When the interactions of $|1\rangle$ with the other states, which, from (13), contribute to b^2, are large enough, the laser associated with α_i is effectively taken out of resonance.

IV. CYCLIC CONFIGURATION

For the branch configurations investigated above, as with cascade configurations, it is possible to find transformations which make the static Hamiltonian elements real and positive and which remove the phases of the oscillating fields. In this section we consider a cyclic configuration, an example of which is shown in Fig. 2, in which the number of lasers is equal to the number of levels. For such cases we find, interestingly, that the relative phases of the lasers of different frequencies do have a critical effect.

The cyclic configuration of Fig. 2 can be seen as an extension of the three-laser four-level cascade configuration achieved by means of a fourth laser coupling states $|4\rangle$ and $|1\rangle$. An analytic solution for a four-level cyclic configuration has been obtained previously.[8] Here we consider two different configurations which display the important physical effect of the relative laser phases.

Interaction terms of the laboratory-frame Hamiltonian involve products of the type $\alpha_j\,|j\rangle\langle j-1\,|$ for $j=1,2,3,4$, where the state $|j-1\rangle$ for $j=1$ represents the state $|4\rangle$.

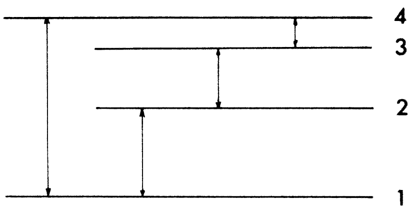

FIG. 2. Example of a four-level system in resonant interaction with four lasers in a cyclic configuration. Levels 1 and 4 are coupled by the laser associated with Rabi frequency α_1, levels 1 and 2 by laser α_2, levels 2 and 3 by laser α_3 and levels 3 and 4 by laser α_4.

Transforming the phases of the states $|j\rangle$ to give $|j\rangle\exp(i\beta_j)$ can only give cancellation of the phase factor in $\alpha_j=|\alpha_j|\exp(i\theta_j)$ for all values of j if a particular relationship $\theta_1+\theta_2+\theta_3+\theta_4=2n\pi$ holds between the phases of the α_j. To keep the discussion as general as possible, so far we do not assume such a relationship, which means that we can only make three of the α_j real and positive, and leave α_1, say, to include an arbitrary phase factor $\exp(i\beta)$, i.e.,

$$\alpha_j=|\alpha_j| \quad \text{for } j\neq 1,\quad \alpha_1=|\alpha_1|\exp(i\beta)\ . \tag{19}$$

The laboratory-frame Hamiltonian is then

$$\mathscr{H}_L=\sum_j |j\rangle\langle j|E_j+\sum_j 2\cos(\omega_j t+\phi_j)\times(\alpha_j|j\rangle\langle j-1|+\text{H.c.})\ , \tag{20}$$

where α_j,ω_j,ϕ_j pertain to the laser beam j which couples $|j\rangle$ and $|j-1\rangle$. Given the resonance conditions $\omega_j=E_j-E_{j-1}$ for $j\neq 1$ and $\omega_1=E_4-E_1$, the transformation

$$T=\exp\left[i\sum_j |j\rangle\langle j|(E_j t+\theta_j)\right] \tag{21}$$

will produce a static Hamiltonian $\mathscr{H}$ when the Bloch-Siegert terms are ignored, which will have zero diagonal elements. We find that if the phase of the α_1 laser is selected so that

$$\phi_1=\phi_2+\phi_3+\phi_4-\beta\ , \tag{22}$$

then we can choose values of θ_j such that $\theta_j-\theta_{j-1}=\phi_j$ for $j\neq 1$ and $\theta_4-\theta_1=\phi_1+\beta$, which makes all the elements of $\mathscr{H}$ real and positive. We then have

$$\mathscr{H}=\sum_j |\alpha_j|(|j\rangle\langle j-1|+|j-1\rangle\langle j|)\ , \tag{23}$$

which gives

$$\mathscr{H}^2=\sum_j [(|\alpha_j|^2+|\alpha_{j+1}|^2)|j\rangle\langle j| + |\alpha_j\alpha_{j-1}|(|j\rangle\langle j-2|+\text{H.c.})]\ , \tag{24}$$

where $|j-2\rangle$ represents the states $|3\rangle$ and $|4\rangle$ when $j=1$ and $j=2$, respectively, α_{j+1} represents α_1 when $j=4$, and so on in cyclic order. From (23) and (24) we obtain

$$\mathscr{H}^3=\sum_j (|\alpha_{j-1}\alpha_{j-2}\alpha_{j+1}|+|\alpha_{j-1}^2\alpha_j|+|\alpha_j^3| + |\alpha_j\alpha_{j+1}^2|)(|j\rangle\langle j-1|+\text{H.c.})\ . \tag{25}$$

It follows that if we adjust one of the Rabi frequencies, e.g., α_1, so that

$$|\alpha_{j-1}\alpha_{j+1}|=|\alpha_j\alpha_{j-2}|\ , \tag{26}$$

which implies $|\alpha_4\alpha_2|=|\alpha_1\alpha_3|$, we have

$$\mathscr{H}^3=\sum_j (|\alpha_{j-2}|^2+|\alpha_{j-1}|^2+|\alpha_j|^2+|\alpha_{j+1}|^2)\times|\alpha_j|(|j\rangle\langle j-1|+\text{H.c.}) =b^2\mathscr{H}\ , \tag{27}$$

where

$$b^2=\sum_j |\alpha_j|^2\ . \tag{28}$$

Thus with this choice of phase and intensity of the laser producing α_1, the cyclic configuration of Fig. 2 is analytically solvable with the solution given by (6). We note that a special case of (26) in which the moduli of all the Rabi frequencies are equal is also a particular case of the configuration of Stettler *et al.*[8] From (6) we find the following transition probabilities:

$$P_{21}=\alpha_2^2 b^{-2}\sin^2[b(t-t_0)]\ , \tag{29}$$

$$P_{41}=|\alpha_1|^2 b^{-2}\sin^2[b(t-t_0)]\ , \tag{30}$$

$$P_{31}=(|\alpha_1|\alpha_4+\alpha_3\alpha_2)^2 b^{-4}\{\cos[b(t-t_0)]-1\}^2\ , \tag{31}$$

$$P_{11}=\{1-2(|\alpha_1|^2+\alpha_2^2)b^{-2}\sin^2[\tfrac{1}{2}b(t-t_0)]\}^2\ . \tag{32}$$

From (31) it can be seen that in the special case $|\alpha_1|=\alpha_4$ and $\alpha_2=\alpha_3$, if all the population is initially in state $|1\rangle$, say, then at a later stage all of the population can be found in state $|3\rangle$, the state not directly coupled to $|1\rangle$.

To investigate the influence of the laser phases, we return to Eq. (20) describing the cyclic configuration of Fig. 2, but now alter the phase of the laser associated with α_1 by π, so the phase condition (22) is replaced by

$$\phi_1=\pi+\phi_2+\phi_3+\phi_4-\beta\ . \tag{33}$$

With this condition, the values of θ_j in (21) can be chosen so that $\theta_j-\theta_{j-1}=\phi_j$ for $j\neq 1$ and $\theta_4-\theta_1=\phi_1+\beta-\pi$, which makes all the elements of $\mathscr{H}$ real and positive except for $\mathscr{H}_{14}$ and $\mathscr{H}_{41}$, which are now real and negative, i.e., $\alpha_1=-|\alpha_1|$, so (23) is replaced by

$$\mathscr{H}=\sum_j \alpha_j(|j\rangle\langle j-1|+\text{H.c.})\ , \tag{34}$$

where α_j is real and positive for $j\neq 1$, and α_1 is negative.

Remembering that the state $|j-2\rangle$ is the same state as $|j+2\rangle$, we obtain

$$\mathscr{H}^2=\sum_j (\alpha_j^2+\alpha_{j+1}^2)|j\rangle\langle j| + \sum_j (\alpha_j\alpha_{j-1}+\alpha_{j+1}\alpha_{j+2})|j\rangle\langle j+2|\ . \tag{35}$$

We see that if $\alpha_j^2+\alpha_{j+1}^2$ is independent of j, then the first term of (35) becomes proportional to the unit operator. Also, if

$$\alpha_j\alpha_{j-1}=-\alpha_{j+1}\alpha_{j+2} \quad (36)$$

for all j, then the second term vanishes. Because α_1 is negative and the other three α_j are positive, we can indeed satisfy both these conditions. This requires the two conditions

$$\alpha_4=\alpha_2,\quad \alpha_1=-\alpha_3\ . \quad (37)$$

With these conditions satisfied, we obtain

$$\mathscr{H}^2=(\alpha_2^2+\alpha_3^2)I\ , \quad (38)$$

which is the simplest form of recurrence relation (3) with $k^2=\alpha_2^2+\alpha_3^2$. The solution for $\overline{U}(t,t_0)$ can be found by substitution into (4) and the various transition probabilities follow. The amplitude that an atom remains in any energy state is given by the cosine term of (4). The interesting feature of the solution is that because $\overline{U}(t,t_0)$ contains no terms of higher order than $\mathscr{H}$, only single-quantum transitions can occur, the amplitudes of which are determined by (34) and the second term of (4). For example, if an atom is initially in state $|1\rangle$ the amplitude that it can be found in $|3\rangle$ is zero at all times. This is in agreement with the result found by Deng[7] for a different physical situation, but which, in the appropriate limit, is mathematically equivalent to the case studied here.

A special case of the above is where, in addition to (37), we also have $\alpha_2=\alpha_3$, so that the magnitudes of all the Rabi frequencies are equal. Equality of all Rabi-frequency magnitudes also defines a special case of the first cyclic configuration of this section, for which the phase relation is (22), and in which case there is a complete transfer of population from state $|1\rangle$ to state $|3\rangle$ at particular times. This clearly shows the important effect of the laser phase on the system. Physically, the only difference between the above two equal Rabi-frequency-magnitude cases is a phase shift of π in one of the laser beams, which has the effect of changing a complete double-quantum population transfer to a zero double-quantum transfer.

V. CONCLUSION

In this paper we have found an analytical solution to the atomic time development when $n-1$ lasers interact resonantly with an n-level atom in a branch configuration, as opposed to the more usually studied cascade configuration. We have assumed that the Rabi frequencies are much greater than the damping rates, so the solutions obtained are applicable for at least a few cycles. We also solved the time evolution of a cyclic configuration involving four lasers interacting with a four-level atom. The interaction Hamiltonian was kept general, but various relationships between the Rabi frequencies and between the laser phases were needed to obtain analytical solutions. For such n-level n-laser interactions the relative phases of the lasers have an important influence on the atomic evolution, and we found for the four-level case that a shift in phase of π of one of the lasers could change the situation from one of maximum double-quantum transfer to one in which the amplitudes for double-quantum transitions are zero. The latter case may be interpreted as complete destructive interference of the amplitudes for a double-quantum transition associated with two different pathways.

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