Triple optical resonance*

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A general method is given to determine the steady-state level populations for N-level systems interacting with N-1 strong resonant laser beams. For a three-level system it reproduces recent results of Whitley and Stroud. For a four-level system it leads to simple conditions for the population inversion between the highest and the lowest level. However, the population of the highest level can never exceed 0.5.

The purpose of this Comment is to give a straightforward mathematical scheme for calculating the steady-state density matrices of multilevel systems interacting with high-intensity laser beams. This problem was recently solved by Whitley and Stroud[1] (WS) for a three-level system. Our method for a three-level system gives equivalent results to those obtained in WS, Sec. VI, by the so-called "rate equations for the dressed states" and is so simple that it can easily be applied to many-level system.

Let us consider an N-level atomic system interacting with $N-1$ resonant intense laser beams. Following WS we take into account only those radiative losses which are due to spontaneous transitions between the N selected levels of the atomic system. We assume also, as was done in Sec. VI of WS, that the decay rates are small as compared to the Rabi frequencies of resonant transitions. The rotating-wave approximation is used for the description of interaction of all laser beams. Unlike the method used in WS, our method of finding the steady-state density matrix of the atomic system employs semiclassical approximation which is fully justified for intense laser beams.[2]

Under those assumptions the semiclassical Hamiltonian describing the interaction of the N-level atomic system with $N-1$ laser beams has the form

$$H(t)=\sum_{m=1}^{N} E_m b_m^\dagger b_m + \frac{1}{2}\sum_{m=1}^{N-1}(\hbar\Omega_m b_{m+1}^\dagger b_m e^{-i\omega_m t}+\text{H.c.}), \tag{1}$$

where b_m ($b_m^\dagger$) annihilates (creates) the atom in the mth unperturbed state $|m\rangle$ of energy E_m, ω_m is the frequency of the mth laser beam, and Ω_m is the Rabi frequency.[3]

This Hamiltonian can be transformed to a time-independent form H by the following unitary transformation:

$$U(t)=\exp\left[-it\left(\sum_{m=1}^{N}\sum_{k=1}^{m-1}\omega_k b_m^\dagger b_m\right)\right]. \tag{2}$$

This transformation leads from the Schrödinger picture to what will be called the rotating-wave picture.

The Liouville–von Neumann equation for the atomic density matrix $\rho(t)$ in our case can be written in the form

$$ih\frac{d}{dt}\rho(t)=(L+\Lambda)\rho(t), \tag{3}$$

where the Liouville superoperator[4] L describes the evolution of the density matrix due to the coherent interaction between the atomic system and the laser beams. It has the form

$$L\rho(t)=[H,\rho(t)], \tag{4}$$

where H is the time-independent interaction Hamiltonian in the rotating-wave picture.

The superoperator[4] Λ describes radiative losses. For the three-level system Λ was calculated from QED in WS. Its matrix form is given by their Eq. (3.11) as this part of the matrix A which contains only the decay constants. The general framework presented in WS can be used to construct Λ for multilevel systems. It is worth noting that for any number of levels the trace of the matrix $\rho(t)$ is preserved in time if Λ in Eq. (3) is obtained according to WS. The matrix form of Λ for a four-level system will be given later.

The steady-state solution ρ_s of Eq. (3) obeys the equation

$$(L+\Lambda)\rho_s=0, \tag{5}$$

which for an N-level system can be written as a set of N^2 homogeneous linear equations for all matrix elements $(\rho_s)_{ik}$ of the density matrix ρ_s. The density matrix ρ_s, therefore, can be viewed as an

eigenvector of the matrix $(L+\Lambda)$ belonging to the eigenvalue zero.

We will solve Eq. (5) in the lowest-order perturbation theory with respect to Λ.

To this end we first solve the zero-order equation

$$L\rho_s^{(0)} \equiv [H, \rho_s^{(0)}] = 0 , \tag{6}$$

whose solutions describe stationary states of the atomic system in the absence of radiative losses. There are exactly N such states each corresponding to an eigenvector of the Hamiltonian. Let us denote normalized eigenvectors of H belonging to eigenvalues λ_k by $|\lambda_k\rangle$ $(k=1,2,\ldots,N)$. The Hamiltonian matrix H is tridiagonal; this enables one to find[5] explicit expressions for eigenvectors even for an arbitrary number of levels. The solutions of Eq. (6) can be written in the form

$$\rho_s^{(0)} = \rho(\lambda_k) \equiv |\lambda_k\rangle\langle\lambda_k| . \tag{7}$$

Since there are N such density matrices, all belonging to the same eigenvalue zero of L, our problem falls into the category of degenerate perturbation theory.[6]

The N-fold degeneracy is removed by the perturbation term Λ. To find the correct linear combination of matrices $\rho(\lambda_k)$,

$$\rho_s = \sum_{k=1}^{N} a_k \rho(\lambda_k) , \tag{8}$$

we must diagonalize the $N\times N$ matrix Λ_{kl} defined by the set of matrix elements of the superoperator Λ evaluated between zeroth order density matrices $\rho(\lambda_k)$ and $\rho(\lambda_l)$,

$$\Lambda_{kl} \equiv (\rho(\lambda_k)|\Lambda\rho(\lambda_l)) \equiv \sum_{ijmn} \rho_{ij}(\lambda_k)\Lambda(ij,mn)\rho_{mn}(\lambda_l) . \tag{9}$$

Actually, it is not even necessary to find all the eigenvectors of Λ_{kl}. Since we are interested in the steady-state solution ρ_s, we must only find the eigenvector of the matrix Λ_{kl} which belongs to the eigenvalue zero. The components a_k of this eigenvector, subject to the normalization condition $\sum_{k=1}^{N} a_k = 1$, give the coefficients of the linear combination (8).

In the three-level case this procedure reproduces the results of WS given by their equations (6.2). It was shown numerically in WS that these results, derived under the assumption that the decay rates are much smaller than the Rabi frequencies, approximate very well the exact results, already when the Rabi frequencies are only a few times larger than the decay rates. We take this as an indication that this is the region of applicability of our method for any number of levels.

We will now briefly sketch the derivation and the results for the four-level case without detuning.

The eigenvalues of the interaction Hamiltonian in the rotating-wave picture are

$$\begin{aligned} \lambda_1 &= -\lambda_2 = \tfrac{1}{2}(c_1^2+c_2^2+c_3^2+\sqrt{\Delta})^{1/2} , \\ \lambda_3 &= -\lambda_4 = \tfrac{1}{2}(c_1^2+c_2^2+c_3^2-\sqrt{\Delta})^{1/2} , \end{aligned} \tag{10}$$

where

$$\Delta = (c_1^2+c_2^2+c_3^2)^2 - 4c_1^2c_3^2 , \tag{11}$$

$$c_k = \tfrac{1}{2}|\Omega_k| . \tag{12}$$

The components, $v_m(\lambda_k) \equiv \langle m|\lambda_k\rangle$, of the normalized eigenvectors of the Hamiltonian H in the representation of unperturbed atomic states are, for the first two eigenvalues,

$$\begin{aligned} v_1(\lambda_1) &= v_1(\lambda_2) = (1/2\sqrt{\Delta})[\Delta+\sqrt{\Delta}\,(c_1^2-c_2^2-c_3^2)]^{1/2} , \\ v_2(\lambda_1) &= -v_2(\lambda_2) = -(1/2\sqrt{\Delta})[\Delta+\sqrt{\Delta}\,(c_1^2+c_2^2-c_3^2)]^{1/2} , \\ v_3(\lambda_1) &= v_3(\lambda_2) = (1/2\sqrt{\Delta})[\Delta+\sqrt{\Delta}\,(-c_1^2+c_2^2+c_3^2)]^{1/2} , \\ v_4(\lambda_1) &= -v_4(\lambda_2) = (1/2\sqrt{\Delta})[\Delta+\sqrt{\Delta}\,(-c_1^2-c_2^2+c_3^2)]^{1/2} , \end{aligned} \tag{13}$$

and, for the last two eigenvalues,

$$\begin{aligned} v_1(\lambda_3) &= v_1(\lambda_4) = (1/2\sqrt{\Delta})[\Delta-\sqrt{\Delta}\,(c_1^2-c_2^2-c_3^2)]^{1/2} , \\ v_2(\lambda_3) &= -v_2(\lambda_4) = -(1/2\sqrt{\Delta})[\Delta-\sqrt{\Delta}\,(c_1^2+c_2^2-c_3^2)]^{1/2} , \\ v_3(\lambda_3) &= v_3(\lambda_4) = (1/2\sqrt{\Delta})[\Delta-\sqrt{\Delta}\,(-c_1^2+c_2^2+c_3^2)]^{1/2} , \\ v_4(\lambda_3) &= -v_4(\lambda_4) = (1/2\sqrt{\Delta}\,[\Delta-\sqrt{\Delta}\,(-c_1^2-c_2^2+c_3^2)]^{1/2} . \end{aligned} \tag{14}$$

The components of four zero-order solutions for the density matrices are

$$\rho_{ij}(\lambda_k) = v_i(\lambda_k)v_j(\lambda_k) \quad (k=1,2,3,4) . \tag{15}$$

In close analogy with WS we assume that the perturbation superoperator Λ has only the following matrix elements $\Lambda(ij,mn)$ different from zero; diagonal elements:

$$\begin{aligned} &\Lambda(12,12) = -\Gamma_{21}, \quad \Lambda(13,13) = -\Gamma_{32} , \\ &\Lambda(14,14) = -\Gamma_{41}-\Gamma_{43}, \quad \Lambda(21,21) = -\Gamma_{21} , \\ &\Lambda(22,22) = -2\Gamma_{21} , \quad \Lambda(23,23) = -\Gamma_{32}-\Gamma_{21}, \\ &\Lambda(24,24) = -\Gamma_{41}-\Gamma_{43}-\Gamma_{21}, \quad \Lambda(31,31) = -\Gamma_{32} , \\ &\Lambda(32,32) = -\Gamma_{32}-\Gamma_{21}, \quad \Lambda(33,33) = -2\Gamma_{32}, \\ &\Lambda(34,34) = -\Gamma_{41}-\Gamma_{43}-\Gamma_{32}, \\ &\Lambda(41,41) = -\Gamma_{41}-\Gamma_{43} , \\ &\Lambda(42,42) = -\Gamma_{41}-\Gamma_{43}-\Gamma_{21} , \\ &\Lambda(43,43) = -\Gamma_{41}-\Gamma_{43}-\Gamma_{32} , \\ &\Lambda(44,44) = -2\Gamma_{41}-2\Gamma_{43} , \end{aligned}$$

off-diagonal elements:

$$\Lambda(22,11)=2\Gamma_{21}, \quad \Lambda(33,22)=2\Gamma_{32},$$
$$\Lambda(44,11)=2\Gamma_{41}, \quad \Lambda(44,33)=2\Gamma_{43};$$

where Γ_{ij} are decay rates from the ith unperturbed atomic level to the jth level.

The 4×4 matrix Λ_{kl}, essential for our calculations, has the form

$$\|\Lambda_{kl}\|=\begin{Vmatrix} A & C & D & D \\ C & A & D & D \\ E & E & B & F \\ E & E & F & B \end{Vmatrix}, \tag{16}$$

where

$$A=-2\{\Gamma_{43}[v_4^2(\lambda_1)-v_4^2(\lambda_1)v_3^2(\lambda_1)]+\Gamma_{41}[v_4^2(\lambda_1)-v_4^2(\lambda_1)v_1^2(\lambda_1)]+\Gamma_{32}[v_3^2(\lambda_1)-v_3^2(\lambda_1)v_2^2(\lambda_1)]$$
$$+\Gamma_{21}[v_2^2(\lambda_1)-v_2^2(\lambda_1)v_1^2(\lambda_1)]\}, \tag{17}$$
$$C=2[\Gamma_{43}v_4^2(\lambda_1)v_3^2(\lambda_1)+\Gamma_{41}v_4^2(\lambda_1)v_1^2(\lambda_1)+\Gamma_{32}v_3^2(\lambda_1)v_2^2(\lambda_1)+\Gamma_{21}v_2^2(\lambda_1)v_1^2(\lambda_1)], \tag{18}$$
$$D=2[\Gamma_{43}v_4^2(\lambda_3)v_3^2(\lambda_1)+\Gamma_{41}v_4^2(\lambda_3)v_1^2(\lambda_1)+\Gamma_{32}v_3^2(\lambda_3)v_2^2(\lambda_1)+\Gamma_{21}v_2^2(\lambda_3)v_1^2(\lambda_1)]. \tag{19}$$

Matrix elements B, E, and F are obtained from A, C, and D by interchanging everywhere λ_1 and λ_3.

The normalized right eigenvector of the matrix (16) belonging to the eigenvalue zero has the following components:

$$a_1=a_2=D(2D-A-C)^{-1}, \tag{20a}$$
$$a_3=a_4=-\tfrac{1}{2}(A+C)(2D-A-C)^{-1}. \tag{20b}$$

Steady-state level populations as functions of decay rates and Rabi frequencies are

$$(\rho_s)_{44}=\mathfrak{N}[(\Gamma_{43}+\Gamma_{41}+\Gamma_{32})c_2^2c_3^2+\Gamma_{21}c_2^2(c_1^2+c_2^2)], \tag{21a}$$
$$(\rho_s)_{33}=\mathfrak{N}[\Gamma_{43}c_2^2(c_2^2+c_3^2)+(\Gamma_{41}+\Gamma_{32}+\Gamma_{21})c_1^2c_2^2], \tag{21b}$$
$$(\rho_s)_{22}=\mathfrak{N}\{\Gamma_{43}c_2^2(c_1^2+c_2^2)+(\Gamma_{41}+\Gamma_{32})[(c_1^2-c_3^2)^2+c_1^2c_2^2]$$
$$+\Gamma_{21}c_2^2c_3^2\}, \tag{21c}$$
$$(\rho_s)_{11}=\mathfrak{N}\{\Gamma_{43}c_1^2c_2^2+(\Gamma_{41}+\Gamma_{32})[(c_1^2-c_3^2)^2+c_2^2c_3^2]$$
$$+\Gamma_{21}c_2^2(c_2^2+c_3^2)\}, \tag{21d}$$

where

$$\mathfrak{N}=\tfrac{1}{2}\{(\Gamma_{43}+\Gamma_{21})c_2^2(c_1^2+c_2^2+c_3^2)$$
$$+(\Gamma_{41}+\Gamma_{32})[(c_1^2-c_3^2)^2+c_2^2(c_1^2+c_3^2)]\}^{-1}. \tag{22}$$

From Eqs. (21) we determine the conditions for the population inversion, $(\rho_s)_{44}>(\rho_s)_{11}$, between the highest and the lowest level. There are two nonoverlapping regions of the parameters in which this inversion occurs. They are defined by the following inequalities:

$$(\Gamma_{21}-\Gamma_{43})c_2^2>(\Gamma_{41}+\Gamma_{32})(c_1^2-c_3^2)>0, \tag{23a}$$
$$(\Gamma_{41}+\Gamma_{32})(c_3^2-c_1^2)>(\Gamma_{43}-\Gamma_{21})c_1^2>0. \tag{23b}$$

It seems to us that it is easier to satisfy the condition (23a), because the decay rates are usually lower for higher levels. In this case the Rabi frequency of the highest transition must be necessarily smaller than the Rabi frequency of the first transition.

In contrast with what was found in WS for the three-level atom, the highest level population $(\rho_s)_{44}$ in the four-level atom can never exceed 0.5. This population as a function of all parameters does not attain its maximum value for finite values of Rabi frequencies. In the case when $\Gamma_{43}<\Gamma_{21}$, the condition for $(\rho_s)_{44}$ to exceed 0.25 is

$$-(\Gamma_{21}-\Gamma_{43})/(\Gamma_{41}+\Gamma_{32})<(c_3^2-c_1^2)/c_2^2<1. \tag{24}$$

An interesting situation occurs if the pumping is symmetrical, i.e., when $c_1=c_3$. In that case, the four-level system behaves like two saturated two-level systems, viz., $(\rho_s)_{44}=(\rho_s)_{11}$ and $(\rho_2)_{33}=(\rho_s)_{22}$. The population ratio of these two two-level systems is

$$\frac{(\rho_s)_{44}}{(\rho_s)_{33}}=\frac{\Gamma_{21}c_2^2+(\Gamma_{43}+\Gamma_{41}+\Gamma_{32}+\Gamma_{21})c_1^2}{\Gamma_{43}c_2^2+(\Gamma_{43}+\Gamma_{41}+\Gamma_{32}+\Gamma_{21})c_1^2}, \tag{25}$$

and if $\Gamma_{43}<\Gamma_{21}$ it approaches its upper bound Γ_{21}/Γ_{43} when the inner Rabi frequency increases relative to outer Rabi frequencies. With this optimal arrangement the population of the highest level approaches the value $\frac{1}{2}\Gamma_{21}(\Gamma_{21}+\Gamma_{43})^{-1}$.

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[4]We use the term superoperator to distinguish this object from an operator acting in the Hilbert space of state vectors. The superoperator acts linearly on the density matrices. For a detailed discussion of the Liouville–von Neumann equations for open systems see, for example, F. Haake, in *Quantum Statistics in Optics and Solid-State Physics*, Springer Tracts in Modern Physics (Springer, Berlin, 1973) Vol. 66.

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