PHYSICAL REVIEW A VOLUME 30, NUMBER 5 NOVEMBER 1984

Nonlinear coherence in three-level systems and SU(3) symmetries

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(Received 19 May 1983; revised manuscript received 12 March 1984)

The problem of nonlinear coherence in three-level systems exposed to two monochromatic radiation fields is treated using the unitary symmetries of SU(3). The Hamiltonian and the initial density operator are brought by a series of unitary transformations into a form in which the problem is reduced to a nutation between the two levels connected by the two-photon transition, i.e., to a time-dependent unitary rotation within the proper SU(2) subgroup. The net effect of the two modes in which the photons can combine in the nonlinear transition is shown to vanish gradually as the three levels approach an equidistant pattern. This certifies that two-photon processes can occur in systems which obey SU(3) symmetry, but not in those which obey the symmetry of SU(2). Four independent constants of motion are identified for the process of two-photon nutation in three-level systems.

I. INTRODUCTION

Nonlinear coherence induced in three-level quantum systems by two radiation fields has been studied theoretically by a number of authors.[1–4] They showed that it was possible to introduce a generalized Bloch vector in analogy with the two-level case. The precession of this vector under the combined action of two radiation fields could describe the evolution of coherence in a given three-level system.

An alternative description has been developed for spins $I=1$ in interaction with a single radiation field.[5–7] The Hamiltonian and the equilibrium density operator were expressed in terms of fictitious spin operators and then the coherent time evolution of the density operator was found by solving the Liouville equation. It has been noted[5] that fictitious spin operators represented in the eigenbase of I_z^2 were identical to the generators of SU(3).[8]

Recently, renewed interest has been paid to the problem of nonlinear coherence in three-level systems.[9–13] The new discoveries included constants of motion in the time evolution of the coherence vector and solutions in the case of slight two-photon off resonance. However, in all those works it was assumed that each of the two radiation fields could affect only one of the two transitions of the three-level quantum system.

When one allows, as in the present paper, that each of the two fields can affect both transitions, one obtains the full interaction of the fields with the system. The nonlinear two-photon processes can occur under the effect of both modes in which the photons from the two fields can combine to form a transition. The group theoretical methods used in the present paper provide a clear physical insight into how the two modes compete and give the net result. Besides a quantitative correction to the previous results,[1–4] the full treatment establishes a qualitatively new result. Namely, the previous partial treatments would give a nonvanishing two-photon coherent nutation even in the case of an equidistant three-level system, while the full treatment shows that the effects of the two modes tend to mutually cancel each other as the quantum levels become equidistant. This finding has a fundamental importance since it demonstrates that nonlinear coherence cannot be induced in quantum systems which obey the internal symmetry of SU(2).

The operator method, used throughout the present paper, enables one to find the constants of motion using the commutation relations of the generators of SU(3). Four independent constants of motion can be obtained for the system treated in the present paper. Also, one can demonstrate that the consideration of the commutation relations can be used to rederive the three previously known constants of motion[11] and obtain a new one.

This paper is structured in the following way. Section II is devoted to the time evolution of the coherence induced by two monochromatic radiation fields in a three-level quantum system. In Sec. III we identify the possible constants of motion and present a discussion of the issue. Section IV concludes the paper.

II. TWO-PHOTON COHERENCE

The time evolution of the two-photon coherence induced by two monochromatic radiation fields in a three-level system was found by Brewer and Hahn.[1] From the set of nine coupled differential equations for the components of the generalized Bloch coherence vector, those authors have found the time evolution for each component, one by one. An alternative approach would be to use the operator algebra in order to find the time evolution of the density operator. It then contains the solution for all the components of the Bloch coherence vector at once. The latter procedure has been largely employed in magnetic resonance problems where the spin systems obey the symmetry of SU(2).[14] The only difference for a nonequidistant three-level system is that one should use the algebra of SU(3).[15,16] Recently, the same approach has been used for systems which have the internal symmetry of SU(4).[17,18]

The problem to be treated in this section is depicted in Fig. 1. The levels are denoted by 1, 2, and 3, and it is assumed that the transition 1-3 is resonant for two-photon transitions as indicated. In general, the two photons, of frequencies ω' and ω'', can be combined in two modes which are denoted by I and II. In the previous papers[1–4] only the effect of a single mode was considered.

The Hamiltonian of the system in interaction with two radiation fields can be written as

$$H=H_0-\vec{\mu}\cdot\vec{\mathrm{E}}_1\cos(\omega' t)-\vec{\mu}\cdot\vec{\mathrm{E}}_2\cos(\omega'' t)\ . \tag{2.1}$$

For the sake of convenience we choose to express the Hamiltonian in frequency units. The unperturbed Hamiltonian is represented in its eigenbase $\{\,|\,i\,\rangle\}$ by a diagonal matrix with $(\underline{H}_0)_{ii}=\hbar^{-1}W_i$ $(i=1,2,3)$.

Assuming that $\Delta m=0$, optical transitions can be induced between levels 1 and 2, and between 2 and 3, the Hamiltonian reads in terms of the generators of SU(3)[19]

$$\begin{aligned}H=&-(\omega_{21}+\omega_{32})I_z^{13}-\tfrac{1}{3}(\omega_{21}-\omega_{32})(I_z^{12}-I_z^{23})\\&-2(\alpha_1 I_x^{12}+\beta_1 I_x^{23})\cos(\omega' t)\\&-2(\alpha_2 I_x^{12}+\beta_2 I_x^{23})\cos(\omega'' t)\ ,\end{aligned} \tag{2.2}$$

where $\omega_{ij}=\hbar^{-1}(W_i-W_j)$ and the transition matrix elements are $\alpha_\lambda=(\vec{\mu}\cdot\vec{\mathrm{E}}_\lambda)_{12}=(\vec{\mu}\cdot\vec{\mathrm{E}}_\lambda)_{21}$ and $\beta_\lambda=(\vec{\mu}\cdot\vec{\mathrm{E}}_\lambda)_{23}=(\vec{\mu}\cdot\vec{\mathrm{E}}_\lambda)_{32}$ $(\lambda=1,2)$. The rotating-wave approximation can be made by performing a time-dependent transformation with

$$U_{\mathrm{I}}=\exp\{-i[(\omega'+\omega'')tI_z^{13}+\tfrac{1}{3}(\omega'-\omega'')t(I_z^{12}-I_z^{23})]\} \tag{2.3a}$$

or with

$$U_{\mathrm{II}}=\exp\{-i[(\omega'+\omega'')tI_z^{13}+\tfrac{1}{3}(\omega''-\omega')t(I_z^{12}-I_z^{23})]\}\ . \tag{2.3b}$$

For exact two-photon resonance $\omega_{21}+\omega_{32}=\omega_{31}=\omega'+\omega''$, the effective Hamiltonians are

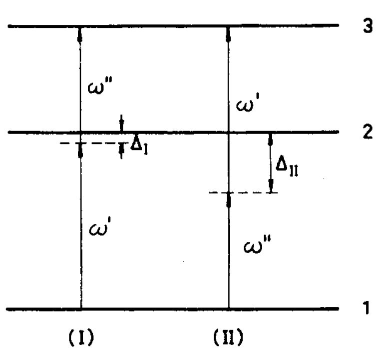

FIG. 1. Energy-level diagram of a three-level system. Two photons of frequencies ω' and ω'' can be combined in two modes, denoted as I and II, in resonant two-photon transitions between levels 1 and 3. The detunings from the intermediate level 2, denoted with Δ_{I} and Δ_{II}, are of the same sign but different in magnitudes.

$$H_{U_{\mathrm{I}}}=-\tfrac{2}{3}\Delta_{\mathrm{I}}(I_z^{12}-I_z^{23})-\alpha_1 I_x^{12}-\beta_2 I_x^{23}\ , \tag{2.4a}$$

$$H_{U_{\mathrm{II}}}=-\tfrac{2}{3}\Delta_{\mathrm{II}}(I_z^{12}-I_z^{23})-\alpha_2 I_x^{12}-\beta_1 I_x^{23}\ , \tag{2.4b}$$

where the oscillating terms have been dropped, and the detunings from the intermediate level 2 denoted as

$$\omega_{21}-\omega'=-(\omega_{32}-\omega'')=\Delta_{\mathrm{I}}\ , \tag{2.5a}$$

$$\omega_{21}-\omega''=-(\omega_{32}-\omega')=\Delta_{\mathrm{II}} \tag{2.5b}$$

for the two modes, respectively.

The two Hamiltonians in Eqs. (2.4) are identical in form. Therefore, we can treat first the effect of a single mode (either I or II) by dropping the indices on Δ, α, and β. The combined effect of both modes is treated subsequently.

A. Effect of a single mode

The Hamiltonian in the rotating-wave approximation has been found to be

$$H_U=-\tfrac{2}{3}\Delta(I_z^{12}-I_z^{23})-\alpha I_x^{12}-\beta I_x^{23}\ . \tag{2.6}$$

Since various terms in this Hamiltonian do not commute, the propagator $\exp(-iH_U t)$ cannot be factored. In order to achieve factorization we first make a unitary transformation with

$$R=e^{-i\theta I_y^{13}}\ , \tag{2.7}$$

where θ is determined by

$$\tan(\tfrac{1}{2}\theta)=\frac{\alpha-\beta}{\alpha+\beta} \tag{2.8}$$

which makes the x operators appear in the transformed Hamiltonian in a symmetrical form

$$H_{UR}=-\tfrac{2}{3}\Delta(I_z^{12}-I_z^{23})-[\tfrac{1}{2}(\alpha^2+\beta^2)]^{1/2}(I_x^{12}+I_x^{23})\ . \tag{2.9}$$

Next, we transform with

$$S=\exp[-2^{-1/2}i\phi(I_y^{12}-I_y^{23})]\ , \tag{2.10}$$

where ϕ is determined by

$$\tan\phi=-\frac{(\alpha^2+\beta^2)^{1/2}}{\Delta}\ . \tag{2.11}$$

The Hamiltonian then reads

$$H_{URS}=-\Lambda(I_z^{12}-I_z^{23})+\Omega I_x^{13}\ , \tag{2.12}$$

where

$$\Lambda=\frac{\Delta}{6}(1+3\cos\phi)-\tfrac{1}{2}(\alpha^2+\beta^2)^{1/2}\sin\phi\ , \tag{2.13a}$$

$$\Omega=\frac{\Delta}{2}(1-\cos\phi)+\tfrac{1}{2}(\alpha^2+\beta^2)^{1/2}\sin\phi\ . \tag{2.13b}$$

The Hamiltonian (2.12) now consists of two commuting terms, and the propagator can be factored.[20]

In order to find the time evolution of the state of the quantum system, one has to choose an initial density operator ρ_0, transform it in the same way as has been used for the Hamiltonian, and then evaluate

$$\rho_{URS}(t)=e^{-iH_{URS}t}(\rho_0)_{URS}e^{iH_{URS}t}\ . \tag{2.14}$$

The time evolution in the original frame can be obtained by transforming $\rho_{URS}(t)$ back by reverse transformations.

The exact solution would include the effects of both, one-photon and two-photon processes. When the intermediate level is close to resonance, the one-photon processes predominate, while for

$$\Delta \gg \alpha,\beta \tag{2.15}$$

the intermediate level is left far off resonance and the two-photon process predominates since it remains to be resonant. Condition (2.15) implies that $\phi\approx 0$ [cf. Eq. (2.11)], and one can approximate S with the unit operator which leaves the density operator unchanged.

As an example we may give the evolution of the system which was initially in level 1 so that

$$\rho_0=\tfrac{1}{3}[I+(I_z^{12}-I_z^{23})]+I_z^{13}\ . \tag{2.16}$$

The time evolution is given by

$$\rho_U(t)=\tfrac{1}{3}[I+(I_z^{12}-I_z^{23})]+I_z^{13}\{1+\cos^2\theta[\cos(\Omega t)-1]\}$$
$$-I_x^{13}\sin\theta\cos\theta[\cos(\Omega t)-1]-I_y^{13}\cos\theta\sin(\Omega t) \tag{2.17}$$

which describes two-photon nutation between levels 1 and 3. The maximum nutation amplitude occurs for $\cos\theta=1$ which requires $\alpha=\beta$. Equation (2.17) predicts the same behavior as the results of the previous papers.[1,3,4]

B. Effect of both modes

In order to evaluate the effect of both modes on the two-photon coherence, one first finds the transformed Hamiltonian for a single mode, say I, with the condition (2.15)

$$H_{(URS)_\mathrm{I}}=-\tfrac{2}{3}\Delta_\mathrm{I}(I_z^{12}-I_z^{23})+\Omega_\mathrm{I}I_x^{13}\ , \tag{2.18}$$

where Ω_I is given by

$$\Omega_\mathrm{I}=-\frac{1}{4}\frac{\alpha_1^2+\beta_2^2}{\Delta_\mathrm{I}}\ . \tag{2.19}$$

Approximating $S_\mathrm{I}^{+}\approx I$, the reverse transformations yield

$$H_{U_\mathrm{I}}=-\tfrac{2}{3}\Delta_\mathrm{I}(I_z^{12}-I_z^{23})+\Omega_\mathrm{I}(I_x^{13}\cos\theta_\mathrm{I}+I_z^{13}\sin\theta_\mathrm{I})\ , \tag{2.20}$$

where θ_I is determined by [cf. Eq. (2.8)]

$$\tan(\tfrac{1}{2}\theta_\mathrm{I})=\frac{\alpha_1-\beta_2}{\alpha_1+\beta_2}\ . \tag{2.21}$$

Next, we transform back with $U_\mathrm{I}^\dagger$, and then forward with U_II. The combined transformation with $U_\mathrm{II}U_\mathrm{I}^\dagger$ replaces Δ_I in the first term of Eq. (2.20) by Δ_II and leaves the second term unchanged. In addition, one retrieves the terms which have been dropped in the rotating-wave approximation for the first mode, so that the Hamiltonian now reads

$$H_U=-\tfrac{2}{3}\Delta_\mathrm{II}(I_z^{12}-I_z^{23})+\Omega_\mathrm{I}(I_x^{13}\cos\theta_\mathrm{I}+I_z^{13}\sin\theta_\mathrm{I})$$
$$-\alpha_2I_x^{12}-\beta_1I_x^{23}\ . \tag{2.22}$$

Following the above procedure for the first mode, we transform with $S_\mathrm{II}R_\mathrm{II}$ and then back with $R_\mathrm{II}^\dagger S_\mathrm{II}^\dagger\approx R_\mathrm{II}^\dagger$. The obtained Hamiltonian is

$$H_U=-\tfrac{2}{3}\Delta_\mathrm{II}(I_z^{12}-I_z^{23})+(\Omega_\mathrm{I}\cos\theta_\mathrm{I}+\Omega_\mathrm{II}\cos\theta_\mathrm{II})I_x^{13}$$
$$+(\Omega_\mathrm{I}\sin\theta_\mathrm{I}+\Omega_\mathrm{II}\sin\theta_\mathrm{II})I_z^{13}\ , \tag{2.23}$$

where Ω_II is given by

$$\Omega_\mathrm{II}=-\frac{1}{4}\frac{\alpha_2^2+\beta_1^2}{\Delta_\mathrm{II}} \tag{2.24}$$

and θ_II is determined by

$$\tan(\tfrac{1}{2}\theta_\mathrm{II})=\frac{\alpha_2-\beta_1}{\alpha_2+\beta_1}\ . \tag{2.25}$$

Since the last two terms in Eq. (2.23) do not commute, the Hamiltonian still has to be transformed with

$$T=e^{-i\xi I_y^{13}}\ , \tag{2.26}$$

where ξ is determined by

$$\tan\xi=\frac{\Omega_\mathrm{I}\sin\theta_\mathrm{I}+\Omega_\mathrm{II}\sin\theta_\mathrm{II}}{\Omega_\mathrm{I}\cos\theta_\mathrm{I}+\Omega_\mathrm{II}\cos\theta_\mathrm{II}}\ . \tag{2.27}$$

The Hamiltonian then reads

$$H_{UT}=-\tfrac{2}{3}\Delta_\mathrm{II}(I_z^{12}-I_z^{23})+\Omega I_x^{13}\ , \tag{2.28}$$

where

$$\Omega=\pm[(\Omega_\mathrm{I}\cos\theta_\mathrm{I}+\Omega_\mathrm{II}\cos\theta_\mathrm{II})^2+(\Omega_\mathrm{I}\sin\theta_\mathrm{I}+\Omega_\mathrm{II}\sin\theta_\mathrm{II})^2]^{1/2}\ . \tag{2.29}$$

The sign corresponds to that of the coefficient of I_x^{13} in Eq. (2.23).

In transforming the initial density operator we note that $\rho_{0U}=\rho_0$ and that the form of T is identical to that of R with θ replaced by ξ. Hence, for the initial density operator taken in the form of Eq. (2.16), one obtains

$$(\rho_0)_{UT}=\tfrac{1}{3}[I+(I_z^{12}-I_z^{23})]+I_z^{13}\cos\xi+1_x^{13}\sin\xi\ . \tag{2.30}$$

Finally, the time evolution of the density operator is given by

$$\rho_{UT}(t)=\tfrac{1}{3}[I+(I_z^{12}-I_z^{23})]+[I_z^{13}\cos(\Omega t)-I_y^{13}\sin(\Omega t)]$$
$$\times\cos\xi+I_x^{13}\sin\xi\ . \tag{2.31}$$

The two-photon nutation occurs with frequency Ω and amplitude $\cos\xi$ in formal analogy with the results for a single mode. A novel feature in the effect of both modes is the possibility of reduced or even zero two-photon nutation amplitude in spite of the presence of two nonvanishing radiation fields. The nutation amplitude for the single mode vanishes only when α and/or β vanish, while in the case of both modes the nutation amplitude vanishes for

$$\Omega_I\cos\theta_I+\Omega_{II}\cos\theta_{II}=\frac{1}{2}\left[\frac{\alpha_1\beta_2}{\Delta_I}+\frac{\alpha_2\beta_1}{\Delta_{II}}\right]=0\ . \quad (2.32)$$

Noting that $\alpha_1\beta_2=\alpha_2\beta_1=\mu_{12}\mu_{23}E_1E_2$, the condition (2.32) reduces to

$$\Delta_I=-\Delta_{II}\ . \quad (2.33)$$

This condition, however, means that the intermediate level 2 is exactly halfway between levels 1 and 3, i.e., that we deal with equidistant levels. This result is not a surprise since three equidistant levels correspond to a system with angular momentum $J=1$ in a dc magnetic field and the three state vectors transform now according to the unitary symmetries of SU(2). No two-photon coherence should be induced in such a system

In order to explain the gradual merging of the consequences of SU(2) and SU(3) symmetries we refer to Fig. 2. The two Δ's are of opposite sign, but not equal in magnitude as in Eq. (2.33). The nutation amplitude is given by

$$\cos\xi=\frac{\mu_{12}\mu_{23}E_1E_2}{2\,|\,\Omega\,|}\left[\frac{1}{\Delta_I}+\frac{1}{\Delta_{II}}\right]. \quad (2.34)$$

When one of the Δ's is much smaller than the other, the effect of one of the modes dominates over that of the other, hence, the latter can be neglected as has been done in the previous works.[1–4,9–12] However, as the Δ's become comparable in their absolute values, the amplitude of the two-photon nutation is reduced according to Eq. (2.34). A more physical insight can be gained by inspection of the nutation frequencies for single modes in Eqs. (2.19) and (2.24). The two frequencies become opposite in sign, hence, one can say that the radiation fields taken in the two modes tend to rotate the density operator simultaneously in two opposite senses. The amplitudes of the single-mode rotations are given by $\cos\theta_I$ and $\cos\theta_{II}$, so that the rate for the combined rotation becomes

$$\Sigma=\Omega_I\cos\theta_I+\Omega_{II}\cos\theta_{II}\ . \quad (2.35)$$

For exactly equidistant levels condition (2.33) holds and the rate for the net rotation Σ vanishes according to Eq. (2.32).

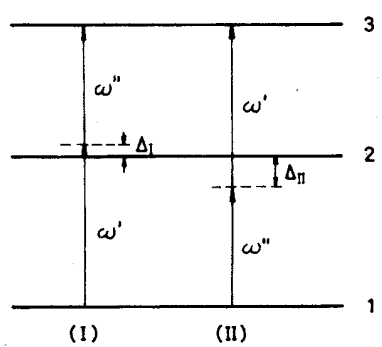

FIG. 2. Energy-level diagram of a system with three almost equidistant levels. The detunings from the intermediate level 2 are slightly different in magnitude and of opposite signs for the two modes in which the photons can combine in the nonlinear transition.

Incidentally, the present results have some bearing on the stationary solutions for the two-photon absorption. Namely, the imaginary part of the third-order nonlinear susceptibility vanishes in the case of two-photon resonance in a system with three equidistant levels when the intermediate level is inbetween the ground and the final level.[21] The present findings explain why the net transition probability for two-photon absorption vanishes in this case.

For nonequidistant three-level systems the maximum nutation amplitude is obtained for

$$\Omega_I\sin\theta_I+\Omega_{II}\sin\theta_{II}=0\ , \quad (2.36)$$

i.e., when the coefficient of I_z^{13} in Eq. (2.23) vanishes and $\xi=0$ [Eq. (2.27)]. The condition (2.36) can be written as

$$\frac{\alpha_1^2-\beta_2^2}{\Delta_I}+\frac{\alpha_2^2-\beta_1^2}{\Delta_{II}}=0\ . \quad (2.37)$$

This relation is satisfied in the trivial case for $\alpha_1=\beta_2$ and $\alpha_2=\beta_1$, which implies $\mu_{12}=\mu_{23}$ and $E_1=E_2$. The condition (2.37) then holds for any Δ_I and Δ_{II}.

For $|\Delta_I| \ll |\Delta_{II}|$ the relation (2.37) is satisfied to a good approximation with $\alpha_1=\beta_2$. In this case the dominant contribution to the two-photon nutation comes from the first mode. In order to achieve the maximum nutation amplitude one has to adjust the field strengths according to $(E_1/E_2)=(\mu_{23}/\mu_{12})$.

When the Δ's are not much different from each other, one has to use Eq. (2.37) in order to adjust the field strengths for maximum nutation amplitude. When nutation amplitude is maximum, the nutation frequency is given by

$$\Omega=\tfrac{1}{2}\mu_{12}\mu_{23}E_1E_2\left[\frac{1}{\Delta_I}+\frac{1}{\Delta_{II}}\right] \quad (2.38)$$

which corresponds to the coefficient of I_x^{13} in Eq. (2.23).

III. CONSTANTS OF MOTION

The constants of motion of a quantum system in interaction with the electromagnetic fields can be grouped in two classes. In the first class one could include the constants of motion which do not depend on the particular form of the Hamiltonian, i.e., do not depend on the driving forces. Such a constant is the trace of the density operator ρ, which, being equal to unity, reflects the total probability of finding the system in all the eigenstates. If the system is in a pure state, there are no further independent constants of motion in this class. However, when the system is in a mixed state, one has $\rho^2\neq\rho$ and there are additional constants of motion. For a two-level system there is one additional constant of motion given by $\mathrm{Tr}\rho^2$. It defines the polarization of the mixed state.[22] For a three-level system the polarization is determined by two independent parameters so that one may expect two corresponding constants of motion. Elgin,[9] and Hioe and Eberly[10] have recently found that these are $\mathrm{Tr}\rho^2$ and $\mathrm{Tr}\rho^3$. The state of polarization of the system cannot be changed whatever the strength, timing, or number of the light pulses. In particular, one cannot drive the system from a statistical mixture into a pure state.[10]

The second class consists of the constants of motion which depend on the particular form of the Hamiltonian. Within the framework of the mathematical treatment adopted in the present paper, these constants of motion can be found using the commutation relations of the generators of SU(3).

The final transformed Hamiltonian, whether for a single mode [Eq. (2.12)] or for both modes [Eq. (2.28)], always has the form

$$H_K = a(I_z^{12} - I_z^{23}) + bI_x^{13} , \tag{3.1}$$

where the index K denotes the total unitary operator used for the transformation. Since the operators $(I_z^{12} - I_z^{23})$ and I_x^{13} mutually commute, they commute also with the Hamiltonian (3.1), and the conserved quantities are

$$Q_1 = \langle (I_z^{12} - I_z^{23}) \rangle_K = \langle K^\dagger (I_z^{12} - I_z^{23}) K \rangle , \tag{3.2}$$

$$Q_2 = \langle I_x^{13} \rangle_K = \langle K^\dagger I_x^{13} K \rangle . \tag{3.3}$$

The equivalence of the two forms on the right-hand side of Eqs. (3.2) and (3.3) comes from the identity

$$\mathrm{Tr}(\rho_K I_p^{ij}) = \mathrm{Tr}(\rho K^\dagger I_p^{ij} K) , \tag{3.4}$$

where the transformed density operator is

$$\rho_K = K \rho K^\dagger . \tag{3.5}$$

Thus, the constants of motion can be expressed in terms of the matrix elements of either the transformed density operator ρ_K or the original density operator ρ.

The operators such as $(I_z^{12} - I_z^{23})^2$ and $(I_p^{13})^2$ also commute with the Hamiltonian. However, they can be expressed as linear combinations of the unit operator I and the operator $(I_z^{12} - I_z^{23})$ and therefore do not provide for new independent constants of motion. Note that the unit operator commutes with the Hamiltonian of any form so that the constant of motion $\langle I \rangle = \mathrm{Tr}\rho$ pertains to the first class discussed above.

Besides the constants of motion Q_1 and Q_2, which are linear in the density matrix elements, one can also find some nonlinear ones. Noting that the propagator $\exp(-iH_K t)$ transforms the generators I_y^{13} and I_z^{13} into each other, one finds that the part of the density operator ρ_K expressed in terms of I_y^{13} and I_z^{13} evolves in time separately from the rest. Since the coefficients of I_p^{ij} in the expansion of ρ_K are given by $2\langle I_p^{ij} \rangle_K$, the corresponding constant of motion can be written as

$$Q_3 = \langle I_y^{13} \rangle_K^2 + \langle I_z^{13} \rangle_K^2 = \langle K^\dagger I_y^{13} K \rangle^2 + \langle K^\dagger I_z^{13} K \rangle^2 . \tag{3.6}$$

Similarly, since the propagator $\exp(-iH_K t)$ transforms the generators I_x^{12}, I_x^{23}, I_y^{12}, and I_y^{23} into each other, the corresponding part of the density operator also evolves as a separate unit. Hence the second nonlinear constant of motion

$$\begin{aligned} Q_4 &= \langle I_x^{12} \rangle_K^2 + \langle I_x^{23} \rangle_K^2 + \langle I_y^{12} \rangle_K^2 + \langle I_y^{23} \rangle_K^2 \\ &= \langle K^\dagger I_x^{12} K \rangle^2 + \langle K^\dagger I_x^{23} K \rangle^2 + \langle K^\dagger I_y^{12} K \rangle^2 + \langle K^\dagger I_y^{23} K \rangle^2 . \end{aligned} \tag{3.7}$$

For the sake of completeness it is necessary to relate the above results to those of Hioe and Eberly.[11] To that end we have to transform the Hamiltonian of Eq. (2.6) in a different way than in Sec. II A. The unitary operator

$$L = e^{-i\eta I_y^{13}} \tag{3.8}$$

transforms I_x^{12} and I_x^{23} into each other. Choosing η according to

$$\tan\eta = -\frac{\beta}{\alpha} \tag{3.9}$$

one can make the coefficient of I_x^{23} in the transformed Hamiltonian to vanish. The result is

$$\begin{aligned} H_{UL} &= -\tfrac{2}{3}\Delta(I_z^{12} - I_z^{23}) - (\alpha^2+\beta^2)^{1/2} I_x^{1/2} \\ &= \frac{\Delta}{3}(I_z^{13} - I_z^{32}) - \Delta I_z^{12} - (\alpha^2+\beta^2)^{1/2} I_x^{12} . \end{aligned} \tag{3.10}$$

Following the above outlined procedures, one can identify a linear constant of motion

$$C_1 = \langle (I_z^{13} - I_z^{32}) \rangle_L = \langle L^\dagger (I_z^{13} - I_z^{32}) L \rangle \tag{3.11}$$

and two nonlinear ones

$$\begin{aligned} C_2 &= \langle I_x^{12} \rangle_L^2 + \langle I_y^{12} \rangle_L^2 + \langle I_z^{12} \rangle_L^2 \\ &= \langle L^\dagger I_x^{12} L \rangle^2 + \langle L^\dagger I_y^{12} L \rangle^2 + \langle L^\dagger I_z^{12} L \rangle^2 , \end{aligned} \tag{3.12}$$

$$\begin{aligned} C_3 &= \langle I_x^{23} \rangle_L^2 + \langle I_x^{13} \rangle_L^2 + \langle I_y^{23} \rangle_L^2 + \langle I_y^{13} \rangle_L^2 \\ &= \langle L^\dagger I_x^{23} L \rangle^2 + \langle L^\dagger I_x^{13} L \rangle^2 + \langle L^\dagger I_y^{23} L \rangle^2 + \langle L^\dagger I_y^{13} L \rangle^2 . \end{aligned} \tag{3.13}$$

The constants of motion C_2, C_3, and C_1 are equivalent to the ones in Ref. 11, Eqs. (19a), (19b), and (19c), respectively. Note that the orthogonal transformation in Eqs. (9)–(11) of Ref. 11, which was found due to a remarkable intuition of Hioe and Eberly, is equivalent to an overall unitary transformation of all the generators of SU(3) by the inverse of (3.8).[23]

Besides C_1, there should be still one linear constant of motion in analogy with the existence of Q_1 and Q_2 above. It can easily be found if the Hamiltonian in Eq. (3.10) is transformed further by the unitary operator

$$P = e^{-i\xi I_y^{12}} , \tag{3.14}$$

where ξ is determined by

$$\tan\xi = \frac{\Delta}{(\alpha^2+\beta^2)^{1/2}} . \tag{3.15}$$

One obtains

$$H_{ULP} = \frac{\Delta}{3}(I_z^{13} - I_z^{32}) - (\Delta^2+\alpha^2+\beta^2)^{1/2} I_x^{12} \tag{3.16}$$

which is formally similar to the Hamiltonian in Eq. (3.1). The second linear constant of motion is

$$C_4 = \langle I_x^{12} \rangle_{LP} = \langle L^\dagger P^\dagger I_x^{12} P L \rangle . \tag{3.17}$$

This constant of motion has not been found by Hioe and Eberly.[11]

IV. DISCUSSION AND CONCLUSIONS

In the present paper we have shown that the classical problem of two-photon coherence in a three-level system can be solved using the transformation properties of SU(3). The Hamiltonian and the initial density operator have been expressed as linear combinations of the generators whose transformation properties within SU(3) could then be used to perform a series of unitary transformations which brought the Hamiltonian into a form where the three-level problem was reduced to a two-level one. The two-photon nutation appeared as a time-dependent unitary rotation within the corresponding SU(2) subgroup.

The two photons, which take part in the nonlinear transition, can be combined in two modes with unequal resonance offsets from the intermediate level. Taking into account only one of the modes, one finds that both the amplitude and the frequency of two-photon nutation depend on the field strengths and the detuning from the intermediate level. The nutation amplitude becomes maximum when Rabi frequencies for the one-photon nutations are made equal ($\alpha=\beta$). In that case, the two-photon nutation frequency is proportional to the square of the one-photon Rabi frequency and inversely proportional to the resonance detuning from the intermediate level. Since the detuning can be given either positive or negative values, the corresponding rotation within SU(2) subgroup occurs in one of the two opposite senses. This revelation has been a clue to the understanding of the absence of two-photon nutation, and, correspondingly, two-photon steady-state absorption, in systems with three equidistant levels. In this case the fields in the two modes tend to perform rotations at equal rates but in opposite senses so that the net rotation vanishes. In terms of unitary symmetries one can say that nonlinear two-photon processes can occur in three-level systems which obey SU(3) symmetry, but not in those which obey the symmetry of SU(2).

We have shown that the constants of motion, which depend on the particular form of the Hamiltonian, can be obtained using the commutation relations of the generators of SU(3). Four constants of motion have been identified, two of them linear and two nonlinear in the density matrix elements. The obtained constants of motion are not unique since any function of them is also a conserved quantity. However, we have always found four independent constants of motion for the system treated in the present paper.

We have also shown that the Hamiltonian can be transformed in two different ways, which lead to two different sets of four constants of motion. The two sets present different, but equally valid pictures for the conserved quantities in the time evolution of the system. The first three constants of the set $\{C_i\}$ have been identified as the ones found previously by Hioe and Eberly,[11] while the fourth is a new one.

Finally, one should discuss the extension of the present findings to the case when the field amplitudes and frequencies are time-dependent parameters, i.e., when the fields are not monochromatic. Hioe and Eberly[11] have correctly noted that their orthogonal transformation holds not only for monochromatic fields, but also in the case when the two amplitudes have the same time dependence so that one may write $\alpha(t)=a\Omega_0(t)$ and $\beta(t)=b\Omega_0(t)$. The same holds for the treatment adopted in the present paper. This can readily be seen by inspection of Eq. (3.9) which gives constant η under the above-stated conditions for $\alpha(t)$ and $\beta(t)$. Hence the unitary operator (3.8) remains time independent. However, for a general time dependence of the field parameters, one would have to look for the Fourier components and reduce the problem to the interaction of the quantum system with a multitude of monochromatic fields.

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