Linear and nonlinear constants of motion for two-photon processes in three-level systems

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Explicit expressions for the constants of motion and the complete solution for the time evolution of the density matrix of a three-level system excited by two laser fields whose amplitudes are sinusoidally modulated and have a phase difference of $\pi/2$ are presented.

Following the discovery by Hioe and Eberly[1] of a set of nonlinear constants of motion for three-level systems excited by two lasers, Gottlieb[2] noted two independent *linear* constants of motion when the fields are sinusoidally amplitude modulated and have a phase difference of $\pi/2$. He also noted a remarkable fact that the time-evolution matrix in that case becomes time independent, so the equations of motion may be solved in terms of its eigenvalues.

It is the purpose of this note to present the following results: (1) to clarify the difference in the nature of linear and nonlinear constants of motion in these works, and (2) to present a new formulation of the problem which can be used very simply to write down explicitly two linear constants of motion as well as the complete solution for the time evolution of the density matrix for the general case when neither the frequency of the modulated laser field amplitudes nor the one-photon detuning vanishes. The latter results were not given by Gottlieb as it would be considerably more cumbersome to arrive at them using his method.

Linear constants of motion can be seen to arise from the following consideration. If the Hamiltonian $\hat{H}$ of an N-level system excited by a laser field is *time independent* (after the rotating wave approximation is taken) as in the case when the laser field amplitudes and frequencies are constant, then it readily follows from the Liouville equation governing the dynamical evolution of the density matrix $\hat{\rho}(t)$ given by

$$i\hbar\partial\hat{\rho}(t)/\partial t=[\hat{H},\hat{\rho}(t)] \ , \tag{1}$$

that we have immediately N linear constants of motion given by

$$\mathrm{tr}[\hat{\rho}(t)\hat{H}^j]=\mathrm{tr}[\hat{\rho}(0)\hat{H}^j]=K_j, \quad j=0,1,2,\ldots,N-1 \ . \tag{2}$$

The N constants of motion are independent of each other unless $\hat{H}$ is of rank less than N. For $j\geq N$, $\hat{H}^j$ can always be expressed in terms of some linear combination of $\hat{H}$ of lower powers by Hamilton-Cayley theorem and hence need not be included.

That K_j given by Eq. (2) are (linear) constants of motion is true regardless of the form of $\hat{H}$, so long as $\hat{H}$ is time independent. For example, consideration of whether the system has equal detunings or at multiphoton resonance is irrelevant.

If the Hamiltonian of the system is time dependent (after the rotating wave approximation is taken), then the quantities given by Eq. (2) are no longer constants of motion. The most general set of constants of motion in this case, for incident laser fields of arbitrary strength, time dependence, and resonance character, are[3]

$$\mathrm{tr}\,\hat{\rho}(t)^j=\mathrm{tr}\,\hat{\rho}(0)^j=C_j, \quad j=1,2,\ldots, \tag{3}$$

of which N of them are generally independent unless $\hat{\rho}$ is of rank less than N. The constants of motion are nonlinear in $\hat{\rho}$ except for the trivial one when $j=1$. Equation (3) resembles the equation giving the Casimir operators $\hat{\mathscr{C}}_n$ in terms of the generators $\hat{P}_{ij}$ of U(N) algebra:

$$\hat{\mathscr{C}}_n=\sum_{i,j,\ldots,l}\hat{P}_{ij}\hat{P}_{jk}\cdots\hat{P}_{li} \ , \tag{3'}$$

where each term in the summation is a product for n factors, $n=1,2,\ldots$. Moreover, for appropriately chosen $\hat{P}_{ij}$, we can represent $\rho_{mn}(t)=\langle\hat{P}_{nm}\rangle\equiv\mathrm{tr}[\hat{\rho}(t)\hat{P}_{nm}]$. It is important to recognize, however, that Eq. (3) is not a direct consequence of Eq. (3′), as $\langle\hat{P}_{ij}\hat{P}_{ji}\rangle$ cannot be equated to $\langle\hat{P}_{ij}\rangle\langle\hat{P}_{ji}\rangle$ generally. The origin of Eq. (3) can be found in the unitarity of $\hat{\rho}(t)$.

If the time dependent Hamiltonian assumes certain symmetrical forms, then there may be further subclasses of constants of motion, depending on the Hamiltonian, in addition to the set given by Eq. (3). These were the nonlinear constants of motion quoted in Refs. 1 and 4, in which it should be noted that the detunings and Rabi frequencies are generally time dependent but of specific forms required by the specified symmetries. Two distinct types of symmetry have been discussed[4] depending on the way the subgrouping schemes are exploited:

$$\mathrm{SU}(3)\supset\mathrm{SU}(2)\times\mathrm{U}(1) \text{ or } \mathrm{SU}(3)\supset\mathrm{O}(3) \ .$$

We now consider the three-level systems excited by two laser fields whose frequencies are kept at two-photon resonance and whose amplitudes are time dependent. The laser fields are assumed to consist of two classical fields of amplitudes $\vec{\mathscr{E}}_j(t)$ and frequencies $f_j(t), j=1,2$. Let $\alpha(t)$ and $\beta(t)$ be the half Rabi frequencies defined by

$$\alpha(t)=\hbar^{-1}\vec{d}_{12}\cdot\vec{\mathscr{E}}_1(t), \quad \beta(t)=\hbar^{-1}\vec{d}_{23}\cdot\vec{\mathscr{E}}_2(t) \ ,$$

where $\vec{d}_{12}$ and $\vec{d}_{23}$ are the atomic dipole moments between levels 1 and 2, and 2 and 3. The detunings $\Delta_{j,j+1}(t)$ are defined as usual by $\Delta_{j,j+1}(t)=|\nu_{j,j+1}|-f_j(t)$ where $|\nu_{j,j+1}|$ is the frequency separation betweeen levels j and $j+1$. At two-photon resonance, we have $\Delta_{12}(t)=-\Delta_{23}(t)\equiv\Delta(t)$. The time evolution of the density matrix is given by

$$i\hbar\partial\hat{\rho}(t)/\partial t=[\hat{H}(t),\hat{\rho}(t)] \ , \tag{4}$$

where $\hat{H}(t)$ can be chosen to be

$$\hat{H}(t)=-\hbar\begin{pmatrix}0 & \alpha(t) & 0\\ \alpha(t) & \Delta(t) & \beta(t)\\ 0 & \beta(t) & 0\end{pmatrix} \ . \tag{5}$$

We introduce the unitary matrix

$$\hat{Q}=\hat{Q}^{\dagger}=\frac{1}{\epsilon}\begin{pmatrix}\alpha & 0 & \beta\\ 0 & \epsilon & 0\\ \beta & 0 & -\alpha\end{pmatrix}\;, \tag{6}$$

where $\epsilon=(\alpha^2+\beta^2)^{1/2}$. Defined also

$$\hat{\rho}'=\hat{Q}^{\dagger}\hat{\rho}\hat{Q}\;, \tag{7}$$

and

$$\hat{H}'=\hat{Q}^{\dagger}\hat{H}\hat{Q}\;. \tag{8}$$

For the case considered by Hioe and Eberly,[1] $\alpha(t)$ and $\beta(t)$ have the same time dependence but possibly different amplitudes:

$$\alpha(t)=a\Omega_0(t),\;\;\beta(t)=b\Omega_0(t)\;. \tag{9}$$

Then $\hat{Q}$ is time independent, and Eq. (5) can be written

$$i\hbar\,\partial\hat{\rho}'(t)/\partial t=[\hat{H}'(t),\hat{\rho}'(t)]\;, \tag{10}$$

where

$$\hat{H}'(t)=-\hbar\begin{pmatrix}0 & \epsilon(t) & 0\\ \epsilon(t) & \Delta(t) & 0\\ 0 & 0 & 0\end{pmatrix}\;. \tag{11}$$

The form of $\hat{H}'(t)$ in Eq. (11) suggests that the dynamical space of $\hat{\rho}'$ can be factorized. It can be readily shown that Eq. (10) can be written in the form given by Eq. (15) of Hioe and Eberly[1]

$$d\vec{\mathrm{T}}(t)/dt=\underline{\Lambda}(t)\vec{\mathrm{T}}(t)\;, \tag{12}$$

where $\underline{\Lambda}(t)$ is a block diagonal matrix consisting of three independent matrices of dimensions 3, 4, and 1, and the three independent nonlinear constants of motion follow.

We now consider the case introduced by Gottlieb[2] in which Δ is constant and $\alpha(t)$ and $\beta(t)$ have the forms

$$\alpha(t)=A\cos\left(\omega t+\phi+\frac{\pi}{2}\right),\;\;\beta(t)=A\cos(\omega t+\phi)\;, \tag{13}$$

with common constant amplitude $A=\epsilon=(\alpha^2+\beta^2)^{1/2}$ but differing by phase $\pi/2$ from each other. The unitary matrix $\hat{Q}$ of Eq. (6) is time dependent in this case. A remarkable fact is that Eq. (5) can now be written as

$$i\hbar\,\partial\hat{\rho}'(t)/\partial t=[\hat{H}'',\hat{\rho}'(t)]\;, \tag{14}$$

where $\hat{\rho}'$ is given by Eq. (7) and $\hat{H}''$ is *time independent* given by

$$\hat{H}''=-\hbar\begin{pmatrix}0 & \epsilon & -i\omega\\ \epsilon & \Delta & 0\\ i\omega & 0 & 0\end{pmatrix}\;. \tag{15}$$

From Eq. (2), we can immediately write down two linear constants of motion given by

$$\mathrm{tr}(\hat{\rho}'\hat{H}''^{j})=\mathrm{tr}(\hat{Q}\hat{\rho}'\hat{Q}^{\dagger}\hat{Q}\hat{H}''^{j}\hat{Q}^{\dagger})=\mathrm{tr}(\hat{\rho}\hat{H}_e^{j})=K_j\;,$$

$$j=1,2\;,$$

where

$$\hat{H}_e=\hat{Q}\hat{H}''\hat{Q}^{\dagger}=-\hbar\begin{pmatrix}0 & \alpha(t) & i\omega\\ \alpha(t) & \Delta & \beta(t)\\ -i\omega & \beta(t) & 0\end{pmatrix}\;. \tag{16}$$

More explicitly, we have

$$\alpha u_{12}+\beta u_{23}+\Delta\rho_{22}-\omega v_{13}=\text{const}\;, \tag{17a}$$

$$(\alpha^2+\omega^2)\rho_{11}+(\alpha^2+\beta^2)\rho_{22}+(\beta^2+\omega^2)\rho_{33}+\alpha\beta u_{13}-\omega(\beta v_{12}+\alpha v_{23}-\Delta v_{13})=\text{const}\;, \tag{17b}$$

where

$$u_{jk}=\rho_{jk}+\rho_{kj},\;\;v_{jk}=i(\rho_{jk}-\rho_{kj})\;.$$

Equations (17) reduce to the results given by Gottlieb for the special case $\omega=0$ or $\Delta=0$. The general case expressed by Eqs. (17) was not given by Gottlieb as it would be considerably more cumbersome to arrive at it using his method.

The complete solution for the time evolution of $\hat{\rho}(t)$ can also be written down very readily. Let $\vec{\phi}_n$ and $\vec{\psi}_n$ be the right and left eigenvectors, respectively, corresponding to the eigenvalue λ_n of the Hamiltonian $\hat{H}''$ given by Eq. (15), and let $\phi_n(j),\psi_n(j),j=1,2,3$ denote their components. From Eq. (14), we find

$$\rho'_{jk}=\sum_{n,m,p,q=1}^{3}\rho'_{pq}(0)\psi_n(p)\phi_m(q)\phi_n(j)\psi_m(k)e^{-i\mu_{nm}t}\;, \tag{18}$$

where

$$\mu_{nm}=\hbar^{-1}(\lambda_n-\lambda_m)\;. \tag{19}$$

To express the solution in terms of the original density matrix $\hat{\rho}$, we use Eq. (7):

$$\hat{\rho}=\hat{Q}\hat{\rho}'\hat{Q}^{\dagger}\;, \tag{20}$$

where $\hat{Q}$, $\hat{Q}^{\dagger}$ are given by Eq. (6).

The eigenvalues λ_n and eigenvectors $\phi_n(j)=\psi_n^*(j)$ of the Hamiltonian $\hat{H}''$ can be found very simply. The eigenvalue equation is given by (for $\hbar=1$)

$$\lambda^3+\Delta\lambda^2-(\epsilon^2+\omega^2)\lambda-\Delta\omega^2=0\;. \tag{21}$$

Let us consider the following three cases separately.

(a) The case $\omega=0$.

$$\begin{gathered}\lambda_1=0,\;\;\phi_1(1)=\phi_1(2)=0,\;\;\phi_1(3)=1\;;\\ \lambda_{2,3}=\tfrac{1}{2}[-\Delta\pm(\Delta^2+4\epsilon^2)^{1/2}],\;\;\phi_{2,3}(1)=\frac{\epsilon}{(\epsilon^2+\lambda_{2,3}^2)^{1/2}}\;,\\ \phi_{2,3}(2)=\frac{-\lambda_{2,3}}{(\epsilon^2+\lambda_{2,3}^2)^{1/2}},\;\;\phi_{2,3}(3)=0\;.\end{gathered} \tag{22}$$

(b) The case $\Delta=0$.

$$\begin{gathered}\lambda_1=0,\;\;\phi_1(1)=0,\;\;\phi_1(2)=\frac{\omega}{(\epsilon^2+\omega^2)^{1/2}},\\ \phi_1(3)=\frac{-i\epsilon}{(\epsilon^2+\omega^2)^{1/2}}\;;\\ \lambda_{2,3}=\pm(\epsilon^2+\omega^2)^{1/2},\;\;\phi_{2,3}(1)=\mp\frac{i}{\sqrt{2}}\;,\\ \phi_{2,3}(2)=\frac{-i\epsilon}{[2(\epsilon^2+\omega^2)]^{1/2}},\;\;\phi_{2,3}(3)=\frac{\omega}{[2(\epsilon^2+\omega^2)]^{1/2}}\;.\end{gathered} \tag{23}$$

(c) The general case $\omega\neq0,\Delta\neq0$.

$$\lambda=-\tfrac{1}{3}\Delta+\tfrac{2}{3}[3(\epsilon^2+\omega^2)+\Delta^2]^{1/2}\cos\frac{\theta+2k\pi}{3},\;\;k=0,1,2\;,$$

where

$$\cos\theta = -\frac{\Delta}{2}\frac{[9(\epsilon^2-2\omega^2)+2\Delta^2]}{[3(\epsilon^2+\omega^2)+\Delta^2]^{3/2}} ,$$

$$\phi(1)=\frac{\lambda^2+\Delta\lambda-i\omega(\lambda+\Delta)}{\epsilon(-\lambda+i\omega)}b, \quad \phi(2)=b , \tag{24}$$

$$\phi(3)=-\frac{\lambda^2+\Delta\lambda-\epsilon^2+i\omega(\lambda+\Delta)}{\epsilon(-\lambda+i\omega)}b ,$$

with b chosen such that $|\phi(1)|^2+|\phi(2)|^2+|\phi(3)|^2=1$.

Substitutions of these eigenvalues and eigenvectors into Eqs. (18) and (20) give one of few known explicit analytic results involving excitations of an atomic system by two amplitude modulated laser fields with a nonzero phase difference. Two constants of motion of this problem are given by Eq. (17).

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[1]F. T. Hioe and J. H. Eberly, Phys. Rev. A 25, 2168 (1982).

[2]H. P. W. Gottlieb, Phys. Rev. A 26, 3713 (1982).

[3]F. T. Hioe and J. H. Eberly, Phys. Rev. Lett. 47, 838 (1981).

[4]F. T. Hioe, Phys. Rev. A 28, 879 (1983).