

# Bloch space structure, the qutrit wavefunction and atom–field entanglement in three-level systems

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## A B S T R A C T

We have given a novel formulation of the exact solutions for the lambda, vee and cascade three-level systems where the Hamiltonian of each configuration is expressed in the  $SU(3)$  basis. The solutions are discussed from the perspective of the Bloch equation and the atom–field entanglement scenario. For the semiclassical systems, the Bloch space structure of each configuration is studied by solving the corresponding Bloch equation and it is shown that at resonance, the eight-dimensional Bloch sphere is broken up into two distinct subspaces due to the existence of a pair of quadratic constants. Because of the different structure of the Hamiltonian in the  $SU(3)$  basis, the non-linear constants are found to be distinct for different configurations. We propose a possible representation of the qutrit wavefunction and show its equivalence with the three-level system. Taking the bichromatic cavity modes to be in the coherent state, the amplitudes of all three quantized systems are calculated by developing an Euler angle based dressed state scheme. Finally following the Phoenix–Knight formalism, the interrelation between the atom–field entanglement and population inversion for all configurations is studied and the existence of collapses and revivals of two different types is pointed out for the equidistant cascade system in particular.

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### Keywords:

$SU(3)$  group  
Three-level system  
Bloch equation  
Entanglement

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## 1. Introduction

In the atom–field interaction scenario, the level structure of an atom leads to the prediction of a wide range of experimentally verifiable coherent phenomena. Probably the most notable among them is the observation of the collapse and revival of the Rabi oscillation [1] which unequivocally proves the granular structure of the photon. This phenomenon is indeed a prediction of the Jaynes–Cummings model—an idealized two-level system consists of an atom with two distinct quantized levels interacting with a monochromatic quantized cavity field [2,3]. An immediate extension of the two-level system is the three-level system which is generally classified into lambda, vee and cascade configurations, respectively. Such configurations are in the purview of present studies because they exhibit a rich class of coherent phenomena such as two-photon coherence [4], the double-resonance process [5], three-level super-radiance [6], resonance Raman scattering [7], population trapping [8], trilevel echoes [9], STIRAP [10], quantum jumps [11], the quantum Zeno effect [12], electromagnetically induced transparency [13] etc. From these studies it is quite transparently obvious that increase of the number of levels not only can generate a large number of quantum-optical effects, but also enables us to develop a suitable control mechanism which is extremely important from the experimental point of view. Thus the three-level configuration, the simplest representative of the multi-level system, demands careful inspection from time to time in its own right.

It is well known that the Hamiltonians of the lambda, vee and cascade three-level system types can be described using the atomic basis operator,  $\hat{\sigma}_{\mu\nu} \equiv |\mu\rangle\langle\nu|$  ( $\mu, \nu = 1, 2, 3$ ), where the solution is carried out with two-photon resonance and equal detuning conditions as supplementary conditions [14,15]. Apart from this treatment, another equivalent way of dealing with the three-level system is by the Bloch equation technique, where the eight Bloch vectors are defined on the eight-dimensional Bloch sphere  $S^7$  [16–18]. This method was first initiated by Eberly and Hioe who pointed out the relationship of the three-level system with the  $SU(3)$  group [16]. Their investigation revealed that the quadratic Casimir of the  $SU(3)$  group is manifested through the existence of some non-linear constants, which gives rise to a non-trivial structure of the Bloch space of such a system [18,19]. Later, this result was obtained by solving the pseudo-spin equation [20] and also by the Floquet theory technique [21]. However, in the Bloch equation approach, the lambda, vee and cascade three-level systems are found to be generated by changing the position of the intermediate level  $E_2$ , i.e., the energy levels are arranged as  $E_2 > E_3 > E_1$ ,  $E_1 > E_3 > E_2$  and  $E_3 > E_2 > E_1$ , as shown in Fig. 1 of Ref. [18]. In consequence, irrespectively of the configuration, the interaction term for any one of these three-level systems in the atomic operator basis is given by  $H_I^A = g_1|1\rangle\langle 2| + g_2|2\rangle\langle 3| + h.c.$  ( $A = \Lambda, V$  and  $\Xi$ ). The pitfall of having identical structures of the Hamiltonians for different configurations is that this leads to same set of non-linear constants, which is undesirable because the three-level systems are intrinsically different from one another.

Apart from that, there is another reason for studying the Bloch space structure of three-level systems. In quantum information theory parlance, the qubit is “designated” by various points on the aforesaid unit Bloch sphere [22,23]. A natural extension of the qubit is the qutrit system, which is generally expressed in the computational basis:  $|-\rangle$ ,  $|0\rangle$  and  $|+\rangle$ ; it has drawn wide attention in recent years [24–26]. Although there exist several suggestions for implementing the qutrit system either by treating it as the transverse spatial modes of single photons [27], or through the polarization states of the biphoton field [28,29], significant progress has been made mainly by identifying the qutrit with the three-level system driven by bichromatic laser fields [30]. However, in spite of the significant progress, a proper definition of the qutrit wavefunction and its explicit relation with the Bloch space structure of the three-level system are not available.

Recently a number of quantum-optical systems have come under intense scrutiny with a view to the experimental implementation of various quantum information protocols where the concurrence is considered the most useful dynamical parameter of evolution [31]. The primary reason for such a study is that it is capable of deciphering the nature of the entanglement between two non-local objects which is the key resource in communicating information. For example, in the phenomenon of *entanglement sudden death*, the entanglement between two non-local two-level systems is studied by considering the time evolution of the concurrence [32]. Since the two-level system is essentially an atom–field composite system, it is equally important to understand the dynamics of the entanglement

between its constituents. As regards the two-level system, the correlation between the atom and the field was first considered by Gea-Banacloche in terms of purity [33] and then more systematically by Phoenix and Knight using the notion of entropy [34,35]. The latter authors also argued that the entropy can be used as an operational parameter to quantify the atom-field entanglement which is constrained by the Araki-Lieb limit. Since the three-level configuration is essentially a pump-probe system at heart, if the entanglement between two non-local such systems is realized experimentally, then it is possible to maneuver the entanglement externally. However, the entanglement dynamics of two non-local three-level systems remains an open issue due to the lack of availability of an appropriate definition of the concurrence of the three-level systems [31,36]. Therefore, prior to addressing the problem of the entanglement of two such non-local systems and to develop a consistent theory of the coherent control mechanism for engineering it by means of some driving field, it is worth studying the entanglement scenario of the constituents of a single three-level system following Phoenix and Knight [34].

The primary objective of this paper has two parts. First, we shall study the structure of the Bloch space for all three-level systems while taking different Hamiltonians for different configurations. Unlike in existing treatments [16,18], it is explicitly shown that if the Hamiltonians of the three-level systems are expressed in the  $SU(3)$  basis with the same energy condition, this leads to distinct non-linear constants for different systems. We also give a convenient representation of the qutrit system and discuss its properties. Secondly, after developing an Euler matrix based dressed state formalism, we discuss a systematic scheme for calculating the atomic entropy following Phoenix and Knight [34] and show how the increase of the number of levels influences the atom-field entanglement and the population inversion for all three configurations.

The remainder of the paper is organized as follows. In Section 2 we reconsider the Hamiltonians of the semiclassical lambda, vee and cascade three-level system types and their quantized versions expressed in the  $SU(3)$  basis. Section 3 obtains the solution of the Bloch equations for the semiclassical configurations in order to provide various non-linear constants and gives a possible representation of the wavefunction of the qutrit system. In Section 4, we proceed to obtain the quantized configurations by introducing an Euler angle based dressed state formalism and compare the corresponding atomic entropy with the population inversion for various initial conditions. In Section 5, we numerically study the atomic entropy with the population inversion for various initial conditions and give a possible estimate of the collapse and revival times for the cascade system in the high field approximation. Finally we summarize the main results of the paper and discuss the outlook.

## 2. The models

The time-dependent Hamiltonian  $H(t)$  can be expanded in the basis of the Lie operators as  $H(t) = \sum_{i=1}^N h_i(t)L_i$ , where the  $h_i(t)$  are the linearly independent complex valued functions and the  $L_i$  are the generators which satisfy the Lie algebra  $[L_i, L_j] = i\omega_{ijk}L_k$ . The algebraic structure of the Hamiltonian ensures the existence of a unitary operator  $U(t) = \prod_{i=1}^N \exp[ig_i(t)L_i]$ , where  $g_i(t)$  is the scalar function which is to be evaluated for any specific model. Thus the time-dependent wavefunction can be evaluated either by the unitary method,  $\psi(t) = U(t)\psi(0)$  [37], or by using a suitable dressed state formalism [38,39]. In this section, we develop the Hamiltonian of the semiclassical and quantized three-level systems where the atomic levels are expressed in the  $SU(3)$  basis.

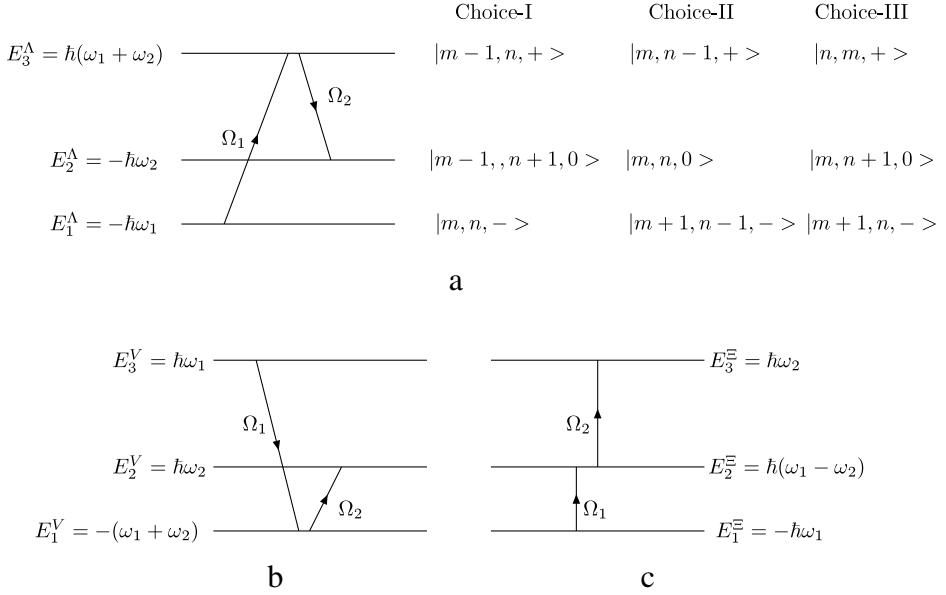
It is customary to consider the three-level system as two two-level systems in the dipole approximation, and the Hamiltonian of the semiclassical lambda system is given by

$$H^\Lambda = \hbar(\omega_1 V_3 + \omega_2 T_3) + \hat{\mathbf{d}}_{13} \cdot \mathbf{E}_1(\Omega_1) + \hat{\mathbf{d}}_{23} \cdot \mathbf{E}_2(\Omega_2), \quad (1)$$

where  $\hat{\mathbf{d}}_{13} = (\mathbf{V}_+ + \mathbf{V}_-)$  and  $\hat{\mathbf{d}}_{23} = (\mathbf{T}_+ + \mathbf{T}_-)$  are the dipole operators representing the transitions  $1 \leftrightarrow 3$  and  $2 \leftrightarrow 3$ , and  $\mathbf{E}_1(\Omega_1) = \mathbf{E}_{1+} + \mathbf{E}_{1-}$  and  $\mathbf{E}_2(\Omega_2) = \mathbf{E}_{2+} + \mathbf{E}_{2-}$  are the bichromatic classical electric fields with mode frequencies  $\Omega_1$  and  $\Omega_2$ , respectively. Similarly we have

$$H^V = \hbar(\omega_1 V_3 + \omega_2 U_3) + \hat{\mathbf{d}}_{13} \cdot \mathbf{E}_1(\Omega_1) + \hat{\mathbf{d}}_{12} \cdot \mathbf{E}_2(\Omega_2), \quad (2)$$

for the vee system, where  $\hat{\mathbf{d}}_{13} = (\mathbf{V}_+ + \mathbf{V}_-)$ ,  $\hat{\mathbf{d}}_{12} = (\mathbf{U}_+ + \mathbf{U}_-)$  are the dipole operators representing transitions  $1 \leftrightarrow 3$  and  $1 \leftrightarrow 2$ , respectively. Finally for the cascade system we have



**Fig. 1.** The energy levels of the lambda, vee and cascade transition types with the energy levels arranged as  $E_3 > E_2 > E_1$ . For each configuration, three choices of the basis states are possible (shown for the lambda system only).

$$H^{\Xi} = \hbar(\omega_1 U_3 + \omega_2 T_3) + \hat{\mathbf{d}}_{12} \cdot \mathbf{E}_1(\Omega_1) + \hat{\mathbf{d}}_{23} \cdot \mathbf{E}_2(\Omega_2), \quad (3)$$

where  $\hat{\mathbf{d}}_{12} = (\mathbf{U}_+ + \mathbf{U}_-)$  and  $\hat{\mathbf{d}}_{23} = (T_+ + T_-)$  are  $1 \leftrightarrow 2$  and  $2 \leftrightarrow 3$  transitions, respectively. In Eqs. (1)–(3),  $\hbar\omega_1 (= -E_1^A)$ ,  $\hbar\omega_2 (= -E_2^A)$  and  $\hbar(\omega_2 + \omega_1) (= E_3^A)$  are the energies of the three levels of the lambda system,  $\hbar\omega_1 (= E_1^V)$ ,  $\hbar\omega_2 (= E_2^V)$  and  $-\hbar(\omega_2 + \omega_1) (= E_3^V)$  are those of the vee system and  $\hbar\omega_1 (= -E_1^{\Xi})$ ,  $\hbar(\omega_1 - \omega_2) (= E_2^{\Xi})$  and  $\hbar\omega_2 (= E_3^{\Xi})$  are the energies of the cascade system, shown in Fig. 1. Here we note that the energy levels of all three configurations satisfy a unique condition  $E_1 < E_2 < E_3$ , and this leads to three distinct interaction Hamiltonians, namely,  $H_I^A = g_1|1\rangle\langle 3| + g_2|3\rangle\langle 2| + h.c.$ ,  $H_I^V = g_1|1\rangle\langle 2| + g_2|1\rangle\langle 3| + h.c.$  and  $H_I^{\Xi} = g_1|1\rangle\langle 2| + g_2|2\rangle\langle 3| + h.c.$ , respectively. These interaction Hamiltonians in the atomic operator basis are precisely identical to the dipole interaction terms in Eqs. (1)–(3) which are expressed in the  $SU(3)$  basis. In Eqs. (1)–(3), we have defined the  $SU(3)$  shift vectors [40],

$$T_{\pm} = \frac{1}{2}(\lambda_1 \pm i\lambda_2), \quad U_{\pm} = \frac{1}{2}(\lambda_6 \pm i\lambda_7), \quad V_{\pm} = \frac{1}{2}(\lambda_4 \pm i\lambda_5),$$

$$T_3 = \lambda_3, \quad V_3 = (\sqrt{3}\lambda_8 + \lambda_3)/2, \quad U_3 = (\sqrt{3}\lambda_8 - \lambda_3)/2, \quad (4)$$

where the Gell-Mann matrices are given by

$$\lambda_1 = \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \lambda_2 = \begin{bmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \lambda_3 = \begin{bmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{bmatrix},$$

$$\lambda_4 = \begin{bmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{bmatrix}, \quad \lambda_5 = \begin{bmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{bmatrix}, \quad \lambda_6 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix},$$

$$\lambda_7 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{bmatrix}, \quad \lambda_8 = \frac{1}{\sqrt{3}} \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{bmatrix}. \quad (5)$$

These matrices follow the commutation and the anti-commutation relations

$$[\lambda_i, \lambda_j] = 2if_{ijk}\lambda_k, \quad \{\lambda_i, \lambda_j\} = \frac{4}{3}\delta_{ij} + 2d_{ijk}\lambda_k, \quad (6)$$

where  $d_{ijk}$  and  $f_{ijk}$  ( $i, j = 1, 2, \dots, 8$ ) represent completely symmetric and completely antisymmetric structure constants, respectively, which characterize the  $SU(3)$  group.

In the rotating wave approximation (RWA), the Hamiltonians of a semiclassical system can be written as [39]

$$H^\Lambda = \hbar(\Omega_1 - \omega_1 - \omega_2)V_3 + \hbar(\Omega_2 - \omega_1 - \omega_2)T_3 + \hbar(\Delta_1^\Lambda V_3 + \Delta_2^\Lambda T_3) + \hbar\kappa_1 V_+ \exp(-i\Omega_1 t) + \hbar\kappa_2 T_+ \exp(-i\Omega_2 t) + h.c., \quad (7)$$

for the lambda system,

$$H^V = \hbar(\Omega_1 - \omega_1 - \omega_2)V_3 + \hbar(\Omega_2 - \omega_1 - \omega_2)U_3 + \hbar(\Delta_1^V V_3 + \Delta_2^V U_3) + \hbar\kappa_1 V_+ \exp(-i\Omega_1 t) + \hbar\kappa_2 U_+ \exp(-i\Omega_2 t) + h.c., \quad (8)$$

for the vee system, and

$$H^\Xi = \hbar(\Omega_1 - \omega_1 + \omega_2)U_3 + \hbar(\Omega_2 + \omega_1 - \omega_2)T_3 + \hbar(\Delta_1^\Xi U_3 + \Delta_2^\Xi T_3) + \hbar\kappa_1 U_+ \exp(-i\Omega_1 t) + \hbar\kappa_2 T_+ \exp(-i\Omega_2 t) + h.c., \quad (9)$$

for the cascade system. In Eqs. (7)–(9),  $\kappa_p$  ( $p = 1, 2$ ) are the coupling parameters and  $\Delta_1^a = (2\omega_1 + \omega_2 - \Omega_1)$  and  $\Delta_2^a = (\omega_1 + 2\omega_2 - \Omega_2)$  for  $a = \Lambda, V$  and  $\Delta_1^\Xi = (2\omega_1 - \omega_2 - \Omega_1)$  and  $\Delta_2^\Xi = (2\omega_2 - \omega_1 - \Omega_2)$  are the respective detuning frequencies.

For the quantized system, the Hamiltonian of the lambda system is given by

$$H^\Lambda = H_0^\Lambda + H_I^\Lambda \quad (10)$$

where

$$H_0^\Lambda = \hbar(\Omega_1 - \omega_1 - \omega_2)V_3 + \hbar(\Omega_2 - \omega_1 - \omega_2)T_3 + \sum_{j=1}^2 \Omega_j a_j^\dagger a_j \quad (11a)$$

$$H_I^\Lambda = \hbar(\Delta_1^\Lambda V_3 + \Delta_2^\Lambda T_3) + \hbar(g_1 V_+ a_1 + g_2 T_+ a_2) + h.c., \quad (11b)$$

with  $a_j^\dagger$  and  $a_j$  ( $j = 1, 2$ ) the creation and annihilation operators of the two cavity modes, the  $g_j$  are the coupling constants and the  $\Omega_j$  are the mode frequencies.

Similarly the Hamiltonian of the vee system is given by

$$H^V = H_0^V + H_I^V \quad (12)$$

where

$$H_0^V = \hbar(\Omega_1 - \omega_1 - \omega_2)V_3 + \hbar(\Omega_2 - \omega_1 - \omega_2)U_3 + \sum_{j=1}^2 \Omega_j a_j^\dagger a_j \quad (13a)$$

$$H_I^V = \hbar(\Delta_1^V V_3 + \Delta_2^V U_3) + \hbar(g_1 V_+ a_1 + g_2 U_+ a_2) + h.c.. \quad (13b)$$

Finally the Hamiltonian of the cascade system in the same approximation is given by

$$H^\Xi = H_0^\Xi + H_I^\Xi \quad (14)$$

where

$$H_0^\Xi = \hbar(\Omega_1 + \omega_2 - \omega_1)U_3 + \hbar(\Omega_2 + \omega_1 - \omega_2)T_3 + \sum_{j=1}^2 \Omega_j a_j^\dagger a_j \quad (15a)$$

$$H_I^\Xi = \hbar(\Delta_1^\Xi U_3 + \Delta_2^\Xi T_3) + \hbar(g_1 U_+ a_1 + g_2 T_+ a_2) + h.c.. \quad (15b)$$

Using the algebra of Gell-Mann matrices it is easy to see that for the lambda and vee systems ( $a = \Lambda$  and  $V$ )

$$[H_0^a, H_I^a] = 0, \quad (16)$$

provided the two-photon resonance condition, namely,

$$\Delta_1^a = -\Delta_2^a, \quad (17)$$

is satisfied. Similarly for the cascade system we must have

$$[H_0^{\mathcal{E}}, H_I^{\mathcal{E}}] = 0, \quad (18)$$

which requires the equal detuning condition, i.e.,

$$\Delta_1^{\mathcal{E}} = \Delta_2^{\mathcal{E}}. \quad (19)$$

Thus the two-photon condition and the equal detuning condition ensure the commutativity of the free and interaction parts of the Hamiltonian, showing that the quantized models are exactly solvable. We now proceed to discuss the Bloch space structure of all three-level configurations by exploring the possible non-linear constants obtained from the semiclassical Hamiltonians in Eqs. (7)–(9) written in the  $SU(3)$  basis with a unique energy condition,  $E_1 < E_2 < E_3$ .

### 3. The Bloch equation and non-linear constants

Let the solution of the Schrödinger equation of the semiclassical three-level system described by the Hamiltonians in Eqs. (7)–(9) be given by

$$\Psi^A(t) = C_-^A(t) |-\rangle + C_0^A(t) |0\rangle + C_+^A(t) |+\rangle, \quad (20)$$

where  $C_-^A$ ,  $C_0^A$  and  $C_+^A$  ( $A = \Lambda, V$  and  $\mathcal{E}$ ) are the amplitudes of the atomic states which satisfy the normalization condition  $|C_-^A|^2 + |C_0^A|^2 + |C_+^A|^2 = 1$ . In Eq. (20) the basis states of the lower, middle and upper atomic states are given by

$$|-\rangle = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}, \quad |0\rangle = \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}, \quad |+\rangle = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}. \quad (21)$$

The exact evaluation of the probability amplitudes enables us to calculate the density matrix defined as

$$\rho^A(t) = |\Psi^A(t)\rangle \otimes \langle \Psi^A(t)|. \quad (22)$$

To start with, we consider the dressed wavefunction of the lambda system obtained by a unitary transformation

$$\tilde{\Psi}^A(t) = U_\Lambda^\dagger(t) \Psi^A(0), \quad (23)$$

where the unitary operator is given by

$$U_\Lambda(t) = \exp \left[ -\frac{i}{3} ((2\Omega_2 - \Omega_1)T_3 + (2\Omega_1 - \Omega_2)V_3)t \right]. \quad (24)$$

The time-independent Hamiltonian of the lambda system described by Eq. (7) is found to be

$$\begin{aligned} \tilde{H}^\Lambda(0) &= -\hbar U_\Lambda^\dagger \dot{U}_\Lambda + U_\Lambda^\dagger H^\Lambda(t) U_\Lambda \\ &= \begin{bmatrix} \frac{1}{3}\hbar(\Delta_1^\Lambda + \Delta_2^\Lambda) & \hbar\kappa_2 & \hbar\kappa_1 \\ \hbar\kappa_2 & \frac{1}{3}\hbar(\Delta_1^\Lambda - 2\Delta_2^\Lambda) & 0 \\ \hbar\kappa_1 & 0 & \frac{1}{3}\hbar(\Delta_2^\Lambda - 2\Delta_1^\Lambda) \end{bmatrix}. \end{aligned} \quad (25)$$

Similarly, the unitary operator of the vee system described by the Hamiltonian in Eq. (8) is

$$U_V(t) = \exp \left[ -\frac{i}{3} ((2\Omega_2 - \Omega_1)U_3 + (2\Omega_1 - \Omega_2)V_3)t \right], \quad (26)$$

and the corresponding time-independent Hamiltonian is given by

$$\tilde{H}^V(0) = \begin{bmatrix} \frac{1}{3}\hbar(2\Delta_1^V - \Delta_2^V) & 0 & \hbar\kappa_1 \\ 0 & \frac{1}{3}\hbar(2\Delta_2^V - \Delta_1^V) & \hbar\kappa_2 \\ \hbar\kappa_1 & \hbar\kappa_2 & -\frac{1}{3}\hbar(\Delta_1^V + \Delta_2^V) \end{bmatrix}. \quad (27)$$

Also the Hamiltonian of the cascade system in Eq. (9) can be made time independent by using the transformation operator

$$U_E(t) = \exp \left[ -\frac{i}{3} ((\Omega_1 + 2\Omega_2)T_3 + (2\Omega_1 + \Omega_2)U_3)t \right] \quad (28)$$

and the corresponding Hamiltonian is

$$\tilde{H}^E(0) = \begin{bmatrix} \frac{1}{3}\hbar(\Delta_1^E + 2\Delta_2^E) & \hbar\kappa_2 & 0 \\ \hbar\kappa_2 & \frac{1}{3}\hbar(\Delta_1^E - \Delta_2^E) & \hbar\kappa_1 \\ 0 & \hbar\kappa_1 & -\frac{1}{3}\hbar(2\Delta_1^E + \Delta_2^E) \end{bmatrix}. \quad (29)$$

Thus we have three distinct Hamiltonians for three different configurations.

To obtain the Bloch equation, we define the generic  $SU(3)$  Bloch vectors

$$S_i^A(t) = \text{Tr}[\rho^A(t)\lambda_i], \quad (30)$$

where  $\rho^A$  is the density matrix given by Eq. (22), which satisfies the Liouville equation

$$\frac{d\rho^A}{dt} = \frac{i}{\hbar}[\rho^A, \tilde{H}^A(0)]. \quad (31)$$

Eq. (30) can be equivalently expressed in terms of a Bloch vector:

$$\rho^A(t) = \frac{1}{3} \left( \mathbf{1} + \frac{3}{2} \sum_{i=1}^8 S_i^A(t)\lambda_i \right). \quad (32)$$

Substituting Eq. (32) in Eq. (31) and making use of the Hamiltonian of the requisite model we obtain the Bloch equation

$$\frac{dS_i^A}{dt} = M_{ij}^A S_j^A, \quad (33)$$

where  $M_{ij}^A$  is the eight-dimensional anti-symmetric matrix. For the Hamiltonian of the lambda system given by Eq. (25), the matrix  $M_{ij}^A$  reads

$$M_{ij}^A = \begin{bmatrix} 0 & \Delta_2^A & 0 & 0 & 0 & 0 & -\kappa_1 & 0 \\ -\Delta_2^A & 0 & 2\kappa_2 & 0 & 0 & -\kappa_1 & 0 & 0 \\ 0 & -2\kappa_2 & 0 & 0 & -\kappa_1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \Delta_1^A & 0 & \kappa_2 & 0 \\ 0 & 0 & \kappa_1 & -\Delta_1^A & 0 & -\kappa_2 & 0 & \sqrt{3}\kappa_1 \\ 0 & \kappa_1 & 0 & 0 & \kappa_2 & 0 & (\Delta_1^A - \Delta_2^A) & 0 \\ \kappa_1 & 0 & 0 & -\kappa_2 & 0 & -(\Delta_1^A - \Delta_2^A) & 0 & 0 \\ 0 & 0 & 0 & 0 & -\sqrt{3}\kappa_1 & 0 & 0 & 0 \end{bmatrix}. \quad (34)$$

Similarly, from the Hamiltonian in Eq. (27) we find the Bloch matrix

$$M_{ij}^V = \begin{bmatrix} 0 & (\Delta_1^V - \Delta_2^V) & 0 & 0 & -\kappa_2 & 0 & -\kappa_1 & 0 \\ -(\Delta_1^V - \Delta_2^V) & 0 & 0 & \kappa_2 & 0 & -\kappa_1 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\kappa_1 & 0 & \kappa_2 & 0 \\ 0 & -\kappa_2 & 0 & 0 & \Delta_1^V & 0 & 0 & 0 \\ \kappa_2 & 0 & \kappa_1 & -\Delta_1^V & 0 & 0 & 0 & \sqrt{3}\kappa_1 \\ 0 & \kappa_1 & 0 & 0 & 0 & 0 & \Delta_2^V & 0 \\ \kappa_1 & 0 & -\kappa_2 & 0 & 0 & -\Delta_2^V & 0 & \sqrt{3}\kappa_2 \\ 0 & 0 & 0 & 0 & -\sqrt{3}\kappa_1 & 0 & -\sqrt{3}\kappa_2 & 0 \end{bmatrix}, \quad (35)$$

for the vee system, and from Eq. (29) we get

$$M_{ij}^{\bar{v}} = \begin{bmatrix} 0 & \Delta_2^{\bar{v}} & 0 & 0 & -\kappa_1 & 0 & 0 & 0 \\ -\Delta_2^{\bar{v}} & 0 & 2\kappa_2 & \kappa_1 & 0 & 0 & 0 & 0 \\ 0 & -2\kappa_2 & 0 & 0 & 0 & 0 & \kappa_1 & 0 \\ 0 & -\kappa_1 & 0 & 0 & (\Delta_1^{\bar{v}} + \Delta_2^{\bar{v}}) & 0 & \kappa_2 & 0 \\ \kappa_1 & 0 & 0 & -(\Delta_1^{\bar{v}} + \Delta_2^{\bar{v}}) & 0 & -\kappa_2 & 0 & 0 \\ 0 & 0 & 0 & 0 & \kappa_2 & 0 & \Delta_1^{\bar{v}} & 0 \\ 0 & 0 & -\kappa_1 & -\kappa_2 & 0 & -\Delta_1^{\bar{v}} & 0 & \sqrt{3}\kappa_1 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{3}\kappa_1 & 0 \end{bmatrix}, \quad (36)$$

for the cascade system.

The algebraic structure of the  $SU(3)$  group allows the existence of a set of quadratic Casimirs which will appear in the form of quadratic constants. To construct them from eight Bloch vectors, we have to search through a total of  $\frac{8!}{(8-n)!n!}$  combinations which will form a tuple. However, because of the large number of such combinations, finding the exact number of such tuples by solving the Bloch equations (34)–(36) is quite onerous. Therefore, to determine all possible non-linear quadratic constants, we have developed a *Mathematica* program to carry out an extensive search and obtain the following results for  $n = 3$  and  $n = 5$  only.

At resonance ( $\Delta_1^A = 0 = \Delta_2^A$ ), for the lambda system the Bloch space is constituted of two parts, one being the 2-sphere  $\mathcal{S}^2$ ,

$$S_1^{A^2}(t) + S_4^{A^2}(t) + S_7^{A^2}(t) = S_1^{A^2}(0) + S_4^{A^2}(0) + S_7^{A^2}(0), \quad (37a)$$

and the other the 4-sphere  $\mathcal{S}^4$ ,

$$\begin{aligned} S_2^{A^2}(t) + S_3^{A^2}(t) + S_5^{A^2}(t) + S_6^{A^2}(t) + S_8^{A^2}(t) \\ = S_2^{A^2}(0) + S_3^{A^2}(0) + S_5^{A^2}(0) + S_6^{A^2}(0) + S_8^{A^2}(0), \end{aligned} \quad (37b)$$

where the  $S_i^A(0)$  are the Bloch vectors at  $t = 0$  which are to be evaluated in terms of the probability amplitudes. On noting the fact that the density matrix can be written as  $\rho^A(t) = U^\dagger(t)\rho^A(0)U(t)$ , the Bloch vector in Eq. (30) becomes

$$S_i^A(0) = \text{Tr}[\rho^A(0)\lambda_i]. \quad (38)$$

Inserting Eq. (5) and the density matrix from Eq. (22) into Eq. (38) at  $t = 0$ , all constants  $S_i^A(0)$  in Eq. (37) can be expressed in terms of probability amplitudes:

$$S_1^{A^2} + S_4^{A^2} + S_7^{A^2} = 4C_-^A(0)^2 C_0^A(0)^2 + 4C_0^A(0)^2 C_+^A(0)^2 \quad (39a)$$















































