Morris-Shore transformation for non-degenerate systems

K. N. Zlatanov, G. S. Vasilev, and N. V. Vitanov

1 Department of Physics, Sofia University, James Bourchier 5 blvd, 1164 Sofia, Bulgaria
2 Institute of Solid State Physics, Bulgarian Academy of Sciences, Tsarigradsko chausse 72, 1784 Sofia, Bulgaria

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The Morris-Shore (MS) transformation is a powerful tool for decomposition of the dynamics of multistate quantum systems to a set of two-state systems and uncoupled single states. It assumes two sets of states wherein any state in the first set can be coupled to any state in the second set but the states within each set are not coupled between themselves. Another important condition is the degeneracy of the states in each set, although all couplings between the states from different sets can be detuned from resonance by the same detuning. The degeneracy condition limits the application of the MS transformation in various physically interesting situations, e.g. in the presence of electric and/or magnetic fields or light shifts, which lift the degeneracy in each set of states, e.g. when these sets comprise the magnetic sublevels of levels with nonzero angular momentum. This paper extends the MS transformation to such situations, in which the states in each of the two sets are nondegenerate. To this end, we develop an alternative way for the derivation of Morris-Shore transformation, which can be applied to non-degenerate sets of states. We present a generalized eigenvalue approach, by which, in the limit of small detunings from degeneracy, we are able to generate an effective Hamiltonian that is dynamically equivalent to the non-degenerate Hamiltonian. The effective Hamiltonian can be mapped to the Morris-Shore basis with a two-step similarity transformation. After the derivation of the general framework, we demonstrate the application of this technique to the popular Λ three-state system, and the four-state tripod, double-Λ and diamond systems. In all of these systems, our formalism allows us to reduce their quantum dynamics to simpler two-state systems even in the presence of various detunings, e.g. generated by external fields of frequency drifts.

I. INTRODUCTION

Coherent control of quantum systems is one of the cornerstones of contemporary quantum physics. Most systems which have been well studied and for which analytical solutions exist consist of only two or three quantum states. In order to make sure that only two or three states are involved in the dynamics the physical system has to be carefully isolated in order to prevent interferences from nearby states with similar energy, which is not always easy to achieve. One of the ways to isolate a system is by large energy separation between ground and excited states. This separation renders the manifolds of ground and excited states to a single pair by largely detuning all other transitions, which diminishes their excitation probability. On the other hand, if the energy separation within a manifold is much smaller than the energy of the coupling field, as is often the case when using angular momentum states or ultrashort laser pulses, such strategy is unreliable. Even with other state isolation techniques, such as light polarization, chirping or light induced energy shifts, additional states often have to be included which adds an extensive complexity to the system.

Multistate systems, by themselves, have many more degrees of freedom and allow the understanding of more complicated intriguing effects. For example, analytical multilevel solutions are essential for the most famous quantum computation algorithms whose building blocks are many-qubits coherent superposition states. Multistate systems play an important role in effects like dark-state polaritons, electromagnetically induced transparency, and multistate population transfer, to name just a few.

Multistate systems are far more difficult to treat than two- and three-state systems as they are described by differential equations of prohibitively high order, unless they can be reduced to simpler systems. One of the most prominent techniques for such reduction is the Morris-Shore (MS) transformation. It is a transformation of the basis vectors in Hilbert space, which reduces two sets of degenerate states to a number of independent two-state systems and residue "dark states", uncoupled from the interaction. The MS transformation has further been generalized to three sets of degenerate states that can reduce the dynamics to independent three-state systems.

The mathematical description of the MS transformation requires the derivation of eigenvalues and eigenvectors of an hermitian matrix, which is not a particularly hard task. There are however a few restrictions on the MS transformation, namely all interactions must share the same time dependence, and also all interactions must be resonant, or equally detuned from the transition frequencies. The last condition implies degeneracy among the states in each of the two sets. While the restriction of same time dependence for the couplings can be met with careful alignment, the condition of degeneracy restricts the applicability of the MS transformation. Indeed, the degeneracy can easily be lifted in the presence of electric and/or magnetic fields or light shifts, for example, when these sets comprise the magnetic sublevels of levels with nonzero angular momentum.
In order to remove this limitation, in this paper we propose an extension of the MS transformation to non-degenerate sets of states. To this end, we propose a new method of obtaining the MS transformation, which allows us to transform a non-degenerate Hamiltonian to a set of independent two-state systems. Essentially we obtain an effective Hamiltonian, which is dynamically equivalent to the non-degenerate Hamiltonian, but whose eigenvalues are much simpler. We further obtain the non-degenerate MS transformation by mapping the diagonalized effective Hamiltonian to the MS basis with a similarity transformation, which can be obtained by the degenerate MS Hamiltonian. We apply our technique to the Λ configuration, which can be obtained by the degenerate MS transformation by mapping the diagonalized effective Hamiltonian, which is dynamically equivalent to the non-degenerate Hamiltonian, but whose eigenvalues are the couplings between the ground and excited states respectively as well as diagonalize the non-degenerate Hamiltonian, but whose eigenvalues are the couplings between the ground and excited states respectively.

The Hamiltonian is a block matrix given by

\[ H = \frac{1}{2} \begin{bmatrix} -\Delta(t)_{g \times g} & V(t)_{g \times e} \\ V(t)^\dagger_{e \times g} & \Delta(t)_{e \times e} \end{bmatrix}. \]

The detuning matrices in Eq. (2) are all diagonal namely,

\[ \Delta(t) = \Delta(t)^{\dagger}I. \]

The time dependent diagonal detunings are defined as the difference of the transition frequency of the system and the frequency of the coupling field

\[ \Delta(t) = (\omega_i - \omega_j) - \omega, \]

where the indices \( i \) and \( j \) run over the excited and ground states respectively. Due to the degeneracy of the system \( \Delta(t) \) is the same for all pairs of coupled states.

The interaction matrix \( V(t) \) is \( g \times e \)-dimensional, whose elements are the couplings between the ground and excited states,

\[ V(t) = \begin{bmatrix} V_{11} & V_{12} & \cdots & V_{1e} \\ V_{21} & V_{22} & \cdots & V_{2e} \\ \vdots & \vdots & \ddots & \vdots \\ V_{g1} & V_{g2} & \cdots & V_{ge} \end{bmatrix}. \]

The idea behind the MS transformation, is to find a unitary matrix \( U \), such that

\[ C^{MS} = UC, \]

\[ H^{MS} = UHU^\dagger = \frac{i}{2} \begin{bmatrix} -\Delta(t) & \Omega(t) \\ \Omega(t)^\dagger & \Delta(t) \end{bmatrix}, \]

where the new interaction matrix \( \Omega(t) \) is now diagonal and the detuning matrix \( \Delta(t) \) of Eq. (3) is left unchanged by the transformation. The consequence of this change of basis is that \( H^{MS} \) can further be rearranged by inspection in a block-diagonal form

\[ H^{MS} = \begin{bmatrix} H_{11}^{MS} & \cdots & \cdots & 0 \\ \vdots & \ddots & \vdots & \vdots \\ \vdots & \vdots & H_n^{MS} & \vdots \\ 0 & \cdots & \cdots & H_{n+1}^{MS} \end{bmatrix}, \]

such that the \( n \) MS Hamiltonians are \( 2 \times 2 \)-dimensional and the \( n + 1 \)-st MS Hamiltonian is a diagonal matrix driving the evolution of uncoupled spectator or "dark" MS states, as illustrated in Fig. II.

The standard Morris-Shore transformation considers a system of \( g \) degenerate ground states coupled to \( e \) degenerate excited states, as described by the time dependent Schrödinger equation in the usual rotating-wave approximation (RWA),

\[ i\hbar \frac{d}{dt} C(t) = H(t)C(t). \]

The Hamiltonian is a block matrix given by

\[ H = \frac{1}{2} \begin{bmatrix} -\Delta(t)^{g \times g} & V(t)^{g \times e} \\ V(t)^\dagger_{e \times g} & \Delta(t)^{e \times e} \end{bmatrix}. \]

The detuning matrices in Eq. (2) are all diagonal namely,

\[ \Delta(t) = \Delta(t)^{\dagger}I. \]

The time dependent diagonal detunings are defined as the difference of the transition frequency of the system and the frequency of the coupling field

\[ \Delta(t) = (\omega_i - \omega_j) - \omega, \]

where the indices \( i \) and \( j \) run over the excited and ground states respectively. Due to the degeneracy of the system
lifts the degeneracy. The new Hamiltonian can then be expressed as

\[
H = \frac{1}{2} \left[ -\Delta(t)_{g\times g} + D_g(t)_{g\times g} \right. \\
\left. V(t)_{g\times e} \right] \\
\left. \Delta(t)_{e\times e} + D_e(t)_{e\times e} \right]
\]

where \( H_0 \) is the degenerate Hamiltonian of Eq. (2) and \( D \) carries the energy shifts.

The complications arising from this non-degeneracy prevent us from using the standard MS transformation by changing basis with \( U \) since the MS Hamiltonian of Eq. (13) reads

\[
H_{MS} = U(H_0 + D)U^\dagger = H_0^{MS} + UDU^\dagger
\]

\[
= \left[ -\Delta(t) + AD_g(t)A^\dagger \right. \\
\left. \Omega \right. \\
\left. \Delta(t) + BD_e(t)B^\dagger \right].
\]

The matrices of the additional term \( UDU^\dagger \), must also be diagonal for the standard MS transformation to remain valid. For the matrix representation of \( U \) in Eq. (5) this is not the case since in general \( AD_g(t)A^\dagger \neq D_g(t) \) and \( BD_e(t)B^\dagger \neq D_e(t) \). This simply means that the MS transformation has to be found by a different procedure than the one used in Section II. An alternative way to achieve the MS transformation is to first diagonalize the Hamiltonian, and then carry a second transformation that will generate the MS Hamiltonian as,

\[
H \xrightarrow{\text{SHS}} W \xrightarrow{\text{PWP}} H^{MS},
\]

where \( W \) is diagonal and the transformation matrices \( S \) and \( P \) satisfy

\[
SHS^\dagger = W = P^\dagger H^{MS}P.
\]

Combined together \( S \) and \( P \) achieve

\[
PSHS^\dagger P^\dagger = UHU^\dagger = H^{MS},
\]

with

\[
U = PS.
\]

In order to utilize the two-step approach we first diagonalize the degenerate Hamiltonian by the matrix of its normalized eigenvectors

\[
SH_{deg}S^\dagger = \Xi,
\]

where

\[
\Xi = \text{diag}(\chi_1, \chi_2, \ldots, \chi_n)
\]

is a diagonal matrix of the eigenvalues. Furthermore, we want to generate an effective Hamiltonian which is dynamically equivalent to the non-degenerate Hamiltonian. The way to achieve this is by taking it as

\[
H_{eff} = S^\dagger BSH_{deg}.
\]
where the matrix \(B\) is to be determined. Now the effective Hamiltonian satisfies the generalized eigenvalue equation

\[
H_{eff} S^\dagger = S^\dagger B \Xi.
\]

If the generalized eigenvalues of the effective Hamiltonian are the same as the eigenvalues of the non-degenerate Hamiltonian they are dynamically equivalent. In other words we want

\[
S H_{eff} S^\dagger = B \Xi = W = R H_{ndeg} R^\dagger
\]

(23)
to hold. Here

\[
W = \text{diag}(\varepsilon_1, \varepsilon_2, ..., \varepsilon_i)
\]

(24)
is the matrix of the eigenvalues of the non-degenerate Hamiltonian of Eq.\((13)\) and \(R\) is composed of its eigenvectors. The matrices \(R\) and \(W\) can be found by any standard diagonalization procedure. In order to find the matrix \(B\) we note that the non-degenerate eigenvalues can be expressed as a series expansion in terms of the energy shifts,

\[
\varepsilon_i = \sum_{k=0}^{\infty} \frac{d^k}{d\delta_i^k} \varepsilon_i |_{\delta_i=0},
\]

(25)
where \(\delta_i\) is the appropriate energy shift that corresponds to the \(i\)-th eigenvalue. Whenever these shifts are small enough we can drop the higher-order terms and only keep the linear expansion

\[
\varepsilon_i \approx \chi_i + \delta_i \kappa_i,
\]

(26)
where

\[
\kappa_i = \frac{d\varepsilon_i}{d\delta_i}|_{\delta_i=0}
\]

(27)
is a function of the control parameters of the Hamiltonian, that is independent of \(\delta_i\). Whenever more than one energy shift is involved in a specific eigenvalue the appropriate vector form of Eq.\((26)\) should be used.

The simplest choice for the matrix \(B\) will be a diagonal form, whose \(i\)-th diagonal element reads

\[
B_i = 1 + \frac{\delta_i \kappa_i}{\chi_i}.
\]

(28)
Whenever an eigenvalue is a zero, it should instead be set to \(\chi_0 = \lim_{p \to 0} p\), as well as the corresponding element in \(B\) reads

\[
B_0 = \lim_{p \to 0} \left(1 + \frac{\delta_0 \kappa_0}{p}\right).
\]

(29)
In this way the generalized eigenvalue evaluates to

\[
\varepsilon_0 = B_0 \chi_0 = \delta_0 \kappa_0.
\]

(30)

Figure 2. Non-degenerate a) \(\Lambda\) system, b) tripod system, c) double \(\Lambda\) system and d) diamond system.

The non-degenerate MS transformation is then obtained by acting with \(U\) on the effective Hamiltonian or with \(P\) on the generalized eigenvalue matrix

\[
H^{MS} = PB \Xi P^\dagger.
\]

(31)
Later on in the text we will use Eq.\((31)\) to generate the MS transformation.

To summarize, our approach has the following sequence of steps. First we find the MS transformation for the degenerate Hamiltonian which gives the matrix \(U\). Then we diagonalize the degenerate MS Hamiltonian to find the map between the diagonal form and the MS basis, which yields the \(P\) matrix and by Eq.\((18)\) the \(S\) matrix. The third step is to diagonalize the non-degenerate Hamiltonian and express its eigenvalue matrix as

\[
W = B \Xi,
\]

(32)
by which we find the \(W\) and \(B\) matrices. Finally the non-degenerate MS Hamiltonian is obtained by Eq.\((31)\).

In the next section we illustrate this two-step generalized eigenvalue approach to the MS transformation with some common systems of high practical significance, namely the \(\Lambda\), the tripod, and the double \(\Lambda\).

**IV. SPECIFIC EXAMPLES**

### A. Lambda system

The simplest case we consider is a \(\Lambda\) system, whose final state has a different energy from the initial state as shown in Fig.\(2\)(a).

The non-degenerate MS transformation is then obtained by acting with \(U\) on the effective Hamiltonian or with \(P\) on the generalized eigenvalue matrix

\[
H^{MS} = PB \Xi P^\dagger.
\]

Later on in the text we will use Eq.\((31)\) to generate the MS transformation.
In order to focus on the MS transformation and to simplify our calculations we will only consider resonant excitation where $\Delta(t) = 0$, although our results can be applied for non-resonant excitation as well. The non-degenerate Hamiltonian reads

$$H_A = \frac{1}{2} \begin{bmatrix} 0 & 0 & \Omega_s & 0 \\ 0 & 2\delta & \Omega_p & 0 \\ \Omega_s & \Omega_p & 0 & 0 \\ 0 & 0 & 0 & \Omega_{rms} \end{bmatrix}, \quad (33)$$

where for the sake of simplicity we have assumed the Rabi frequencies to be real. After we apply the procedure from Section II we find the transformation matrix for the degenerate system to be

$$U = \frac{1}{\Omega_{rms}} \begin{bmatrix} -\Omega_p & \Omega_s & 0 & 0 \\ \Omega_s & \Omega_p & 0 & 0 \\ 0 & 0 & \Omega_{rms} & 0 \\ 0 & 0 & 0 & \Omega_{rms} \end{bmatrix}, \quad (34)$$

where we have introduced the root mean square Rabi frequency

$$\Omega_{rms} = \sqrt{\Omega_p^2 + \Omega_s^2}. \quad (35)$$

The degenerate MS Hamiltonian is then

$$H_A^{MS} = \frac{1}{2} \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & \Omega_{rms} & 0 \\ 0 & \Omega_{rms} & 0 & 0 \\ 0 & 0 & 0 & \Omega_{rms} \end{bmatrix}. \quad (36)$$

Furthermore, its diagonalization reads

$$P^\dagger H_A^{MS} P = \Xi = \frac{1}{2} \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & \Omega_{rms} & 0 \\ 0 & \Omega_{rms} & 0 & 0 \\ 0 & 0 & 0 & \Omega_{rms} \end{bmatrix}, \quad (37)$$

with the matrix $P$ given as

$$P = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & -\frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \\ 0 & \frac{1}{\sqrt{2}} & 0 & -\frac{1}{\sqrt{2}} \end{bmatrix}. \quad (38)$$

The eigenvalues of the non-degenerate Hamiltonian are too cumbersome to be presented here so instead we directly present the result for the generalized eigenvalues. From Eq. (26), Eq. (28) and Eq. (29) we find them to be

$$\Xi = \begin{bmatrix} -\delta & 0 & 0 & 0 \\ 0 & \Omega_{rms} & 0 & 0 \\ 0 & 0 & \Omega_{rms} & 0 \\ 0 & 0 & 0 & \Omega_{rms} \end{bmatrix}. \quad (39)$$

In order to estimate how good our approximation is in Fig. 3 we have compared the bottom corner generalized eigenvalue of Eq. (39) with its corresponding eigenvalue of Eq. (33). As evident from the figure the approximation holds quite well as long as the ratio $\delta/\Omega_{rms}$ is of $O(10^{-2})$

\[ \text{Figure 3. Comparison between the third generalized eigenvalue of Eq. (39) and its corresponding eigenvalue of Eq. (33). The parameters have been set to $\Omega_s = 1.25 T$, $\Omega_p = 1.37 T$, and $\Omega_{rms} = 1.85 T$, where $T$ is the pulse duration for the excitation.} \]

order. The MS Hamiltonian (31) for the non-degenerate system reads

$$H_A^{MS} = \begin{bmatrix} \frac{\delta \Omega_p^2}{\Omega_{rms}} & 0 & 0 \\ 0 & \frac{\delta \Omega_s^2}{\Omega_{rms}} & \frac{\delta \Omega_c^2}{\Omega_{rms}} \\ 0 & \frac{\delta \Omega_p^2}{\Omega_{rms}} & \frac{\delta \Omega_c^2}{\Omega_{rms}} \end{bmatrix}. \quad (40)$$

The $\Lambda$ system provides a dark state uncoupled from the evolution of the other two. Often a third state ground state coupled to the excited state is added forming the tripod system, which we explore next.

**B. Tripod system**

The tripod system is illustrated in Fig. 2(b). We assume that the middle coupling is resonant, while the "plus" and "minus" couplings are detuned with $\mp \delta$ respectively due to the lifted degeneracy. The Hamiltonian of the system reads,

$$H_T = \frac{1}{2} \begin{bmatrix} -\delta & 0 & 0 & \Omega_p \\ 0 & 0 & 0 & \Omega_s \\ 0 & 0 & \delta & \Omega_c \\ \Omega_p & \Omega_s & \Omega_c & 0 \end{bmatrix}. \quad (41)$$

The degenerate MS transformation is carried out by

$$U = \begin{bmatrix} \Omega_p \Omega_s & -\sqrt{\Omega_p^2 + \Omega_s^2} & 0 & 0 \\ \Omega_p & \Omega_s & \sqrt{\Omega_p^2 + \Omega_s^2} & 0 \\ 0 & 0 & \Omega_c & \Omega_s \\ \Omega_p & \Omega_s & \Omega_c & \Omega_p \end{bmatrix}. \quad (42)$$

where

$$\Omega_{rms} = \sqrt{\Omega_p^2 + \Omega_s^2 + \Omega_c^2}. \quad (43)$$
Then the standard MS Hamiltonian is
\[ H_{T|\delta=0}^{MS} = \frac{1}{2} \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \Omega_{rms} \\ 0 & 0 & \Omega_{rms} & 0 \end{bmatrix} \].

Diagonalizing it with the matrix
\[ P = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \\ 0 & 0 & -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \end{bmatrix} \]

leaves us with
\[ P^\dagger H_{T|\delta=0}^{MS} P = \frac{1}{2} \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & -\Omega_{rms} & 0 \\ 0 & 0 & 0 & \Omega_{rms} \end{bmatrix} \].

The generalized eigenvalue matrix reads
\[ B \Xi = \begin{bmatrix} \hat{\Delta}_1 & 0 & 0 & 0 \\ 0 & \hat{\Delta}_2 & 0 & 0 \\ 0 & 0 & \frac{\delta \Omega}{\Omega_{rms}^2} - \frac{\Omega_{rms}}{2} & 0 \\ 0 & 0 & 0 & \frac{\delta \Omega}{\Omega_{rms}^2} + \frac{\Omega_{rms}}{2} \end{bmatrix} \],

where we have substituted
\[ \hat{\Omega}_+ = \Omega_p^2 + \Omega_c^2, \]
\[ \hat{\Omega}_- = \Omega_p^2 - \Omega_c^2, \]
\[ \hat{\Delta}_1 = -\frac{\delta \left( \hat{\Omega}_- - \sqrt{4\Omega_+^2 + 4\Omega_+ \Omega_2^2 + \hat{\Omega}_-^2} \right)}{4\Omega_{rms}^2}, \]
\[ \hat{\Delta}_2 = -\frac{\delta \left( \hat{\Omega}_- + \sqrt{4\Omega_+^2 + 4\Omega_+ \Omega_2^2 + \hat{\Omega}_-^2} \right)}{4\Omega_{rms}^2}. \]

Finally the MS transformation of Eq. (41) reads
\[ H_{T}^{MS} = \begin{bmatrix} \hat{\Delta}_1 & 0 & 0 & 0 \\ 0 & \hat{\Delta}_2 & 0 & 0 \\ 0 & 0 & \frac{\delta \Omega}{\Omega_{rms}^2} - \frac{\Omega_{rms}}{2} & 0 \\ 0 & 0 & 0 & \frac{\delta \Omega}{\Omega_{rms}^2} + \frac{\Omega_{rms}}{2} \end{bmatrix} \].

and the detuning. Thus changing them will change not only the global phase but the superposition as well. This comes as a consequence of the zero eigenvalue of the dark state. However if we instead have a non-zero eigenvalue, whose phase contribution depends on a parameter, that doesn’t affect the superposition, we can control the global phase. For example a close look at Eq. (42) reveals that the second MS state will be a superposition of the first and third state in the original basis, that is independent of \( \Omega_s \). During the evolution of the system, however the state remains unchanged besides a phase shift proportional to the detuning element of Eq. (43). This phase shift depends on \( \Omega_s \) thus giving a control parameter that leaves the superposition intact, while changing the global phase.

C. Double \( \Lambda \)

The final example we discuss involves multiple excited states. The simplest case is the double \( \Lambda \) shown in Fig. 2c), which consists of two ground and two excited states which are non-degenerate. This systems can also represented in a diamond configuration, given in Fig. 2d), as the two are similar. In order to simplify the problem we assume that the direct and cross couplings between the ground states and the excited states are equal,
\[ \Omega_{11} = \Omega_{22} = \Omega_d, \]
\[ \Omega_{12} = \Omega_{21} = \Omega_c, \]
as is the case for \( J = 1/2 \rightarrow J = 1/2 \) transitions.

The Hamiltonian reads
\[ H_{2\Lambda} = \frac{1}{2} \begin{bmatrix} 0 & 0 & \Omega_d & \Omega_c \\ 0 & -\delta & \Omega_c & \Omega_d \\ \Omega_d & \Omega_c & 0 & 0 \\ \Omega_c & \Omega_d & 0 & \delta \end{bmatrix}. \]

Following the procedures from Section 11 we find the MS Hamiltonian to be
\[ H_{2\Lambda|\delta=0}^{MS} = \frac{1}{2} \begin{bmatrix} 0 & 0 & \Omega_- & 0 \\ 0 & 0 & 0 & -\Omega_+ \\ \Omega_- & 0 & 0 & 0 \\ 0 & -\Omega_+ & 0 & 0 \end{bmatrix} \],

where we have taken the shorthand notation
\[ \Omega_+ = \Omega_c + \Omega_d, \]
\[ \Omega_- = \Omega_c - \Omega_d. \]

The transformation matrix to the MS basis is then
\[ U = \begin{bmatrix} \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} & 0 & 0 \\ -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 & 0 \\ 0 & 0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \\ 0 & 0 & -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \end{bmatrix}. \]
The next step is to find the map between the MS Hamiltonian and the diagonal form, which reads

$$P^\dagger H^{MS}_{2\Lambda}\delta_{\pi\sigma}=0P = \frac{1}{2}\begin{bmatrix} -\Omega_+ & 0 & 0 & 0 \\ 0 & \Omega_+ & 0 & 0 \\ 0 & 0 & -\Omega_+ & 0 \\ 0 & 0 & 0 & \Omega_+ \end{bmatrix}, \tag{55}$$

with diagonalization matrix

$$P = \begin{bmatrix} 0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\ \frac{1}{\sqrt{2}} & 0 & 0 & \frac{1}{\sqrt{2}} \\ \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 & 0 \\ 0 & 0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \end{bmatrix}. \tag{56}$$

From Eq.\((\ref{eq:56})\) and Eq.\((\ref{eq:51})\) we find the generalized eigenvalue matrix to be

$$BΞ = \begin{bmatrix} \delta_- - \Omega_+ & 0 & 0 & 0 \\ 0 & \delta_- + \Omega_+ & 0 & 0 \\ 0 & 0 & \delta_- - \Omega_+ & 0 \\ 0 & 0 & 0 & \delta_- + \Omega_+ \end{bmatrix}, \tag{57}$$

where

$$\delta_- = \frac{\delta_+ - \delta_\pi}{4}. \tag{58}$$

The MS Hamiltonian is then

$$H^{MS}_{2\Lambda} = \frac{1}{2}\begin{bmatrix} \delta_- & 0 & 0 & 0 \\ 0 & \delta_- & 0 & -\Omega_+ \\ 0 & -\Omega_+ & \delta_- & 0 \\ 0 & 0 & 0 & \delta_- \end{bmatrix}, \tag{59}$$

which can be transformed to a block-diagonal form with the permutation matrix

$$\pi = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}, \tag{60}$$

that finally leaves us with

$$\pi H^{MS}_{2\Lambda} \pi^T = \frac{1}{2}\begin{bmatrix} \delta_- & 0 & 0 & 0 \\ 0 & \delta_- & 0 & -\Omega_+ \\ 0 & -\Omega_+ & \delta_- & 0 \\ 0 & 0 & 0 & \delta_- \end{bmatrix}. \tag{61}$$

The clear benefit of the MS transformation here is the simple two-state picture of the excitation dynamics. Moreover when a number of states, as well as control parameters are involved, the dynamics of the system is not at all obvious. For example in our current case if we set $\Omega_c = \Omega_d$ we will have no coupling in the upper block of Eq.\((\ref{eq:61})\). Furthermore if we initially set all the population in the MS states corresponding to that block, there will be no excitation of any MS state. This conclusion can’t be drawn from Eq.\((\ref{eq:61})\) directly, but comes easily in the MS basis.

V. DISCUSSION AND CONCLUSION

In this paper, we explored the extension of the MS transformation to sets of non-degenerate states. For this purpose, we developed an approach based on first diagonalizing the non-degenerate Hamiltonian, finding a simpler dynamically equivalent effective Hamiltonian, and finally, transferring to the MS basis with a second similarity transformation by diagonalizing the degenerate MS Hamiltonian.

A key point in our analysis and derivation of the effective Hamiltonian of Eq.\((\ref{eq:21})\) is that its eigenvalues match those of the non-degenerate Hamiltonian with high accuracy. In order to fulfill this condition we require that the effective Hamiltonian satisfies the generalized eigenvalue equation \((\ref{eq:22})\). We assume that the energy shifts just slightly perturb the non-degenerate eigenvalues, which justifies the linear approximation in Eq.\((\ref{eq:26})\). In addition, the approximation holds strongly for $\frac{\delta}{\Omega_{rms}} \sim O(10^{-2})$, which describes well optical transitions where the excitation frequency is greater than a THz and the frequency shift among magnetic sublevels, for example, is of the order of MHz. If this is not the case, higher order terms have to be included into the eigenvalue approximation of the effective Hamiltonian.

We illustrated our concept explicitly with four popular systems, namely the A, tripod, double-A and diamond systems, which have numerous applications in a variety of physical situation. Further applications of our results might be expected in the calculation of state fidelities. When an experimental graph of a readout measurement is compared with a theoretical prediction that assumes degeneracy, a discrepancy is to be expected. The cause is due to a variety of factors, that are often prescribed to experimental imperfections rather than inaccuracy of the theoretical model. We expect that our model will account for such discrepancies between laboratory measured fidelity and theoretical predictions due to negligence of non-degeneracy. The magnitude of this effect remains to be investigated.

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