Excitation transfer in two two-level systems coupled to an oscillator

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Abstract
We consider a generalization of the spin-boson model in which two different two-level systems are coupled to an oscillator, under conditions where the oscillator energy is much less than the two-level system energies, and where the oscillator is highly excited. We find that the two-level system transition energy is shifted, producing a Bloch–Siegert shift in each two-level system similar to what would be obtained if the other were absent. At resonances associated with energy exchange between a two-level system and the oscillator, the level splitting is about the same as would be obtained in the spin-boson model at a Bloch–Siegert resonance. However, there occur resonances associated with the transfer of excitation between one two-level system and the other, an effect not present in the spin-boson model. We use a unitary transformation leading to a rotated system in which terms responsible for the shift and splittings can be identified. The level splittings at the anticrossings associated with both energy exchange and excitation transfer resonances are accounted for with simple two-state models and degenerate perturbation theory using operators that appear in the rotated Hamiltonian.

1. Introduction
The coupled quantum system consisting of a two-level system interacting with a harmonic oscillator provides a model that has been the focus of a large number of studies over recent decades. A reduced (Rabi Hamiltonian) version of the problem was published by Bloch and Siegert in studies of the interaction of a dynamical magnetic field perpendicular to a static magnetic field with a spin system [1], and was later applied to the problem of atoms interacting with an electromagnetic mode [2]. The more complete model (the spin-boson Hamiltonian) was introduced much later by Cohen-Tannoudji and collaborators [3].

At the most basic level, the interaction of the two-level system with the oscillator produces a shift as before; however, the oscillator is only weakly impacted by the two-level system since the two-level energy in this case amounts to a small fraction of the total oscillator energy. In this limit the Bloch–Siegert shift is accurately approximated using an adiabatic model [6], and we have obtained new estimates for the level splitting at the anticrossings [7] using degenerate perturbation theory on a rotated version of the model. In the rotated problem, multiphoton transitions are mediated through a complicated ‘perturbation’ operator, but in the end the level splitting is obtained approximately from a simple two-state model with only first-order coupling. The complicated interactions of the original spin-boson problem, which are responsible for energy exchange of a large number of quanta, are reasonably well...
accounted for through the lowest-order coupling in the rotated version of the problem.

This approach, and the resulting conceptual simplification of the problem, is reasonably general and very powerful. We extended the analysis to the case of a spin-one system coupled to an oscillator [9], and obtained results for the Bloch–Siegert shift in agreement with previous work, as well as new results for the level splitting at the Bloch–Siegert resonances. The accuracy was comparable to that obtained for the spin-boson model, and it seems clear that the approach can be applied systematically to higher-spin generalizations of the spin-boson model as well. We also studied a different generalization of the spin-boson model in which a three-level system is coupled to an oscillator [8]. Technical issues associated with the three-level system made the implementation of the rotation much more challenging; however, in the end we obtained good results for the level shifts, and for the level splitting at the anticrossings, as long as the anticrossing occurred away from other strong resonances which interfere.

In this work, we turn our attention to a different generalization of the spin-boson model in which two different two-level systems are coupled to an oscillator. Our approach is the most useful in the multiphoton regime with a highly excited oscillator. In this case, both two-level systems experience Bloch–Siegert shifts due to their interaction with the oscillator, and anticrossings occur associated with energy exchange between each two-level system and the oscillator. The Bloch–Siegert shift for each two-level system in this case is very close to what would be expected if the other two-level system were absent, and the level splittings at the energy exchange anticrossings are not very different from the single two-level version of the problem. What is new in this problem are anticrossings associated with excitation transfer, in which the excitation from one two-level system is transferred to the other. In the rotated version of this model, we find a spin–spin interaction term which mediates these new excitation transfer transitions. Our attention in this work is then focused on excitation transfer, and we find that a simple two-state model and degenerate perturbation theory leads to reasonably accurate estimates of the level splittings away from other resonances.

This model and related models have been studied in connection with studies of the interaction between two atoms and a laser cavity [10–12], and quantum entanglement [13–17]. In the first area, the focus is on cooperative interactions, coherent dynamics and photon statistics as theoretical issues; in the second area, the focus is quantum control and entanglement of neutral atoms in a cavity, and in an optical trap. Excitation transfer itself as a physical mechanism is perhaps the most important in biophysics [18], but in this case it is mediated by the Coulomb interaction. Coherent excitation transfer is of interest for applications in quantum information studies, as noted above.

Our motivation for this study comes about from the possibility of the development of new physical systems that makes use of excitation transfer through indirect coupling, wherein one might be able to transfer the excitation from one atom or molecule to a distant one simply by driving a lower-energy mode that is common to both. As an unfamiliar and perhaps opaque quantum effect, it should be possible to develop a laboratory demonstration (once the effect becomes better understood); then perhaps subsequently applications may suggest themselves, perhaps in the area of quantum information, spectroscopy or sensor technology. We are also interested in whether this mechanism may already operate in existing physical systems, but without having been recognized yet.

The excitation transfer effect in this model is very weak when \( n \) is large. To study it in a regime in which energy exchange resonances do not interfere, we need to work with low oscillator energy (which maximizes the number of oscillator quanta needed for resonance) and modest oscillator excitation (since the level splitting is inversely proportional to the number of oscillator quanta). The reason why excitation transfer is so weak in this model is studied using a finite-basis approximation; in which we see that destructive interference occurs between the contributions from pathways involving intermediate states. We briefly examine the possibility of reducing or eliminating the destructive interference, in order to make the excitation transfer effect stronger. If the model is modified so that the coupling between the different two-level systems is through conjugate oscillator operators, then some of the destructive interference is removed. If the model is augmented with loss terms that remove the contribution of the lower-energy intermediate states, then the destructive interference is completely eliminated; the excitation transfer effect then becomes much stronger.

2. Model

We consider the model described by the Hamiltonian

\[
\hat{H} = \Delta E_1 \frac{\hat{s}_1}{\hbar} + \Delta E_2 \frac{\hat{s}_2}{\hbar} + \bar{h} \omega_0 \hat{a}^\dagger \hat{a} + U_1 (\hat{a}^\dagger + \hat{a}) \frac{2\hat{s}_1}{\hbar} \\
+ U_2 (\hat{a}^\dagger + \hat{a}) \frac{2\hat{s}_2}{\hbar}.
\]

(1)

There are a pair of two-level systems, with unperturbed transition energies \( \Delta E_1 \) and \( \Delta E_2 \); and an oscillator, with characteristic energy \( \hbar \omega_0 \). The first two-level system interacts linearly with the oscillator, with a coupling strength of \( U_1 \); similarly, the second two-level system also interacts linearly with the oscillator, with a coupling strength of \( U_2 \). The spin operators are defined in terms of the Pauli matrices according to

\[
\hat{s}_j = \frac{\hbar}{2} \vec{\sigma}_j,
\]

(2)

where the superscript denotes which two-level system is referenced. In the multiphoton regime, the characteristic energy of the oscillator is much less than the unperturbed transition energies

\[
\hbar \omega_0 \ll \Delta E_1 \quad \hbar \omega_0 \ll \Delta E_2.
\]

(3)
3. Unitary transformation

As in our previous work on spin-boson problems [7, 19], we find it useful to consider a unitary equivalent Hamiltonian

\[ \hat{H}' = \hat{U} \hat{H} \hat{U}^\dagger, \]

where

\[ \hat{U} = e^{i \hat{H}_0 t / \hbar}, \]

(5)

Since \( \sigma^{(1)} \) commutes with \( \sigma^{(2)} \), the computation is very similar to the single two-level problem. This unitary transform is a straightforward generalization of one used previously in connection with the spin-boson problem [20, 21].

3.1. Dressed Hamiltonian and 'unperturbed' part

We can write the rotated Hamiltonian as

\[ \hat{H}' = \hat{H}_0 + \hat{V}_1 + \hat{V}_2 + \hat{W}_1 + \hat{W}_2 + \hat{V}_{12}. \]

(6)

where the 'unperturbed' part of the rotated Hamiltonian \( \hat{H}' \) is given by

\[ \hat{H}_0 = \sqrt{\Delta E_1^2 + 8 U_1^2 y^2 \frac{\sigma^{(1)}}{\hbar}} + \sqrt{\Delta E_2^2 + 8 U_2 y^2 \frac{\sigma^{(2)}}{\hbar}} + \hbar \omega_0 \hat{a} \hat{a}^\dagger, \]

(7)

where the coordinate \( y \) can be related to the creation and annihilation operators according to

\[ y = \frac{\hat{a} + \hat{a}^\dagger}{\sqrt{2}}. \]

(8)

This is similar to the unperturbed part that we obtained previously in the case of the spin-boson model, where now two dressed two-level terms appear instead of one.

In the spin-boson model, and also in other problems that we have studied, the 'unperturbed' part \( \hat{H}_0 \) of the rotated Hamiltonian \( \hat{H}' \) results in a good approximation for the oscillator and dressed transition energy of the two-level systems. It can be used to develop approximations for resonance conditions as we discuss in the following section. Because of this, we have come to think of \( \hat{H}_0 \) as describing an unperturbed version of the dressed problem in which no interactions occur at level crossings. Viewed in this way, the other terms in the rotated Hamiltonian can be thought of as perturbations.

3.2. Perturbations involved in energy exchange resonances

There are now two primary perturbations \( \hat{V}_1 \) and \( \hat{V}_2 \) (only a single one appears in the spin-boson problem since there is only one two-level system in that model), each of which can be described by the general formula

\[ \hat{V}_j = \frac{i \hbar \omega_0}{2} \left( \frac{\sqrt{\Delta E_j}}{\Delta E_j} \right) \frac{d}{dy} \left( \frac{\sqrt{\Delta E_j}}{\Delta E_j} \right) \frac{2 \sigma^{(j)} y}{\hbar}. \]

(9)

where

\[ \frac{d}{dy} = \frac{\hat{a} - \hat{a}^\dagger}{\sqrt{2}}. \]

(10)

In the multiphoton regime, these terms give rise to level splittings at resonances in which one unit of excitation of a two-level system is exchanged for an odd number of oscillator quanta, as discussed in [7].

3.3. Potential operators

There are two small terms \( \hat{W}_1 \) and \( \hat{W}_2 \), both described through the general formula

\[ \hat{W}_j = \frac{i \hbar \omega_0}{2} \left( \frac{U_j}{\Delta E_j} \right)^2 \left( \frac{2 \sigma^{(j)} y}{\hbar} \right)^2. \]

(11)

Since the square of the spin operator \( \sigma \) is proportional to the identity matrix

\[ \left( \frac{2 \sigma^{(j)} y}{\hbar} \right)^2 = \sigma \frac{2 \sigma^{(j)} y}{\hbar}, \]

(12)

these terms become simple potentials

\[ \hat{W}_j = \frac{i \hbar \omega_0}{2} \left( \frac{U_j}{\Delta E_j} \right)^2 \left( \frac{2 \sigma^{(j)} y}{\hbar} \right)^2. \]

(13)

In the large \( n \) limit which is of interest in this paper, these potentials are very small, and do not contribute in a significant way to either the occurrence of resonances or the level splitting at resonance. We neglect them in what follows.

3.4. Spin–spin interaction

Finally, we find a spin–spin operator \( \hat{V}_{12} \) given by

\[ \hat{V}_{12} = \frac{i \hbar \omega_0}{2} \left( \frac{U_{12}}{\Delta E_1} \right)^2 \left( \frac{2 \sigma^{(1)} y}{\hbar} \right)^2 \left( \frac{2 \sigma^{(2)} y}{\hbar} \right)^2. \]

(14)

This operator has no analog in the spin-boson model. In the multiphoton regime this term contributes to the level splitting associated with excitation transfer (where one unit of excitation in a one two-level system is exchanged for one unit of excitation in the other two-level system, accompanied by the exchange of an even number of oscillator quanta). As this effect is new in this model, it will be the focus of our attention in this work.

4. Energy levels and resonance conditions

The time-independent Schrödinger equation for \( \hat{H}_0 \) is

\[ \left( E + \frac{\hbar \omega_0}{2} \right) \Psi = \frac{\hbar \omega_0}{2} \left( -\frac{d^2}{dy^2} + \frac{\Delta E_1^2}{\Delta E_1} \right) \Psi + \Delta E_2^2 + 8 U_2 y^2 \frac{\sigma^{(2)}}{\hbar} \Psi. \]

(15)

4.1. Product solutions

We can reduce this Schrödinger equation to a purely spatial problem in \( y \) by assuming a product wavefunction for \( \Psi \) of the form

\[ \Psi = u_{m_j}(y)|s, m_j\rangle|s, m_2\rangle, \]

(16)

where \( |s, m_j\rangle \) are spin 1/2 eigenkets (with \( s = 1/2 \) and \( m_j = \pm 1/2 \)). This leads to a one-dimensional Schrödinger equation

\[ \epsilon u(y) = \left[ -\frac{d^2}{dy^2} + v(y) \right] u(y), \]

(17)
where the normalized energy eigenvalue $\epsilon$ is

$$\epsilon = \frac{2E}{\hbar \omega_0} + 1 \quad (18)$$

and where the normalized nonlinear potential $v(y)$ is

$$v(y) = y^2 + \frac{2m_1}{\hbar \omega_0} \sqrt{\Delta E_1^2 + 8U_1^2 y^2} \quad + \frac{2m_2}{\hbar \omega_0} \sqrt{\Delta E_2^2 + 8U_2^2 y^2}. \quad (19)$$

We have suppressed the subscripts $m_j$ on $u(y)$.

4.2. Approximate solutions

In the large-$n$ limit, the oscillator is highly excited and we would not expect the two-level systems to have much impact on the oscillator. In this case, a reasonable approximation for the oscillator is to use eigenfunctions of the simple harmonic oscillator

$$u(y) = \langle y | n \rangle, \quad (20)$$

where $| n \rangle$ are the harmonic oscillator eigenfunctions. Within this approximation, we may write

$$E_{n,m_1,m_2} = \Delta E_1(g_1)m_1 + \Delta E_2(g_2)m_2 + n\hbar \omega_0, \quad (21)$$

where the dressed transition energies $\Delta E_1(g_1)$ and $\Delta E_2(g_2)$ are given by

$$\Delta E_j(g_j) = \Delta E_j \left( n \left( 1 + \frac{8U_j^2 y^2}{\Delta E_j^2} \right) \right). \quad (22)$$

The dimensionless coupling constants $g_1$ and $g_2$ are defined according to

$$g_j = \frac{U_j \sqrt{n}}{\Delta E_j}. \quad (23)$$

We interpret $\Delta E_j(g_j)$ as the ‘dressed’ transition energy of the $j$th two-level system. Numerical solutions for the original Hamiltonian $\tilde{H}$ are well described by this expression away from resonances, similar to what we found in the spin-boson problem [7]. This also allows us to meaningfully label our eigenfunctions $u(y)$ with $n$ (again suppressing the weak $m_1$ and $m_2$ dependence)

$$u(y) = u_n(y). \quad (24)$$

4.3. Energy exchange resonances

Within this approximation scheme, we can develop resonance conditions for energy exchange resonances. For energy exchange between the first two-level system and the oscillator, the resonance condition is

$$\Delta E_1(g_1) = \Delta n \hbar \omega_0 \quad (25)$$

with $\Delta n$ odd. These resonances correspond to the ones we analyzed using a similar approach in the spin-boson model [7]. The level splittings at the associated anticrossings are illustrated in figure 1 under conditions where the energy exchange resonances dominate (open circles). Also shown are approximate results using degenerate perturbation theory based on the eigenfunctions of the rotated $\tilde{H}_0$ problem. In this calculation, one state is chosen in which the first two-level system is excited, the second is in the ground state, and $n$ quanta are present in the oscillator; the other state is chosen so that both two-level systems are in the ground state, and $n + \Delta n$ quanta are in the oscillator. The level splitting at the anticrossings using degenerate perturbation theory is [7]

$$\delta E_{\text{min}} = 2|\langle \Psi_{n,2,-1/2} | \hat{V}_1 | \Psi_{n,\Delta n, -1/2, -1/2} \rangle|. \quad (26)$$

One sees that the level splittings are in reasonable agreement with this approximation. This is similar to what we found previously in the spin-boson model. Moreover, the level splitting found in this example matches the other spin-boson model (obtained by removing the second two-level system) such that they could not be distinguished if plotted together in this figure.

Energy exchange between the second two-level system and oscillator occurs similarly when the resonance condition

$$\Delta E_2(g_2) = \Delta n \hbar \omega_0 \quad (27)$$

is satisfied.

4.4. Excitation transfer resonances

This system supports another kind of resonance that is not present in the spin-boson model. Excitation can be transferred from one two-level system to the other (along with energy exchange with the oscillator), which has motivated us to refer to the associated resonances as ‘excitation transfer’ resonances. Consider the resonance between one state, with energy

$$E_{m_1,-1/2,1/2} = -\frac{1}{2} \Delta E_1(g_1) + \frac{1}{2} \Delta E_2(g_2) + \hbar \omega_0 \mu_0 \quad (28)$$

and another, with energy

$$E_{m_2+\Delta n,1/2,-1/2} = \frac{1}{2} \Delta E_1(g_1) - \frac{1}{2} \Delta E_2(g_2) + \hbar \omega_0 (n_0 + \Delta n). \quad (29)$$

Figure 1. Level splittings associated with energy exchange anticrossings, for a model with $\Delta E_1 = 11\hbar \omega_0$ and $\Delta E_2 = 13\hbar \omega_0$, $n_0 = 10^4$. Solid circles: results of direct numerical calculation of the original $\tilde{H}$ problem; open circles: results from degenerate perturbation theory using eigenfunctions of $\tilde{H}_0$. The dimensionless coupling coefficient $g_2$ is fixed at 0.5 for these calculations. The number of oscillator quanta exchanged $\Delta n$ is indicated for each anticrossing.
levels split at anticrossing when resonances occur. We can compute this splitting in at least two ways: either by direct numerical diagonalization of the full unrotated Hamiltonian $\hat{H}$, or by using degenerate perturbation theory on the rotated Hamiltonian.

5.1. Degenerate perturbation theory

In the vicinity of a level anticrossing that is free of energy exchange resonance disruption, we can investigate the level splittings using a two-state approximation of the form

$$\psi = c_0\phi_0 + c_1\phi_1$$

(32)

$$\phi_0 = u_n(y)|s, -1/2\rangle|s, 1/2\rangle$$

(33)

$$\phi_1 = u_{n+\Delta n}(y)|s, 1/2\rangle|s, -1/2\rangle.$$  

The two-state problem leads to the following characteristic equation for the energy levels

$$E \begin{pmatrix} c_0 \\ c_1 \end{pmatrix} = \begin{pmatrix} E_0 & \langle\phi_1|V_{12}|\phi_0\rangle \\ \langle\phi_1|V_{12}|\phi_0\rangle & E_1 \end{pmatrix}.$$  

(34)

Since the basis states are assumed to be at resonance

$$E_0 = E_1$$

the level splitting is given by

$$\delta E_{\text{min}} = 2|\langle\phi_0|V_{12}|\phi_1\rangle|.$$  

(35)

5.2. Level-splitting estimates

The matrix element that appears here can be written as

$$\langle\phi_0|V_{12}|\phi_1\rangle = 2\hbar\omega_0 \frac{U_1}{\Delta E_1 \Delta E_2} \frac{U_2}{n} I_{12} = \frac{2\hbar\omega_0 g_1 g_2}{n} I_{12},$$  

(36)

where the integral $I_{12}$ is given by

$$I_{12} = \int_{-\infty}^{\infty} u_n(y) \frac{1}{[1 + \frac{8|U_1|^2}{\Delta E_1^2}]^{1/2} + [1 + \frac{8|U_2|^2}{\Delta E_2^2}]^{1/2}} dy.$$  

(37)

We can compute this integral using either numerical wavefunctions obtained by diagonalizing $\hat{H}_0$, or by utilizing the WKB approximation [22].

The level splitting on resonance can be expressed in terms of the integral $I_{12}$ as

$$\delta E_{\text{min}} = \frac{4\hbar\omega_0 g_1 g_2}{n} I_{12}.$$  

(38)

The results of the level splittings from a direct numerical computation using the unrotated $\hat{H}$ and degenerate perturbation theory (using the WKB approximation) are illustrated in figure 3. We can see that the perturbation theory results are in excellent agreement with the exact result. In this example, we have selected large two-level system transition energies relative to the oscillator energy in order to minimize the impact of energy exchange resonances. In addition, we have chosen a moderate value for $n$ (instead of a larger value) to increase the level splitting.

5. Level splittings for excitation transfer resonances

As we observed in the case of a single two-level system, energy levels split at anticrossing when resonances occur. We can...
6. Excitation transfer using a finite-basis expansion

The level splitting associated with an excitation transfer resonance is a weak effect in this model when \( n \) is large. We can better understand the slow dynamics of the excitation transfer by making use of a finite-basis expansion. One can see from this kind of calculation that destructive interference between different pathways produces a very small second-order coupling between initial and final states. If this destructive interference can be broken, then the second-order coupling is greatly increased.

6.1. Finite-basis approximation

Indirect coupling between initial and final states associated with an excitation transfer process can be analyzed in the weak coupling limit through the use of a finite-basis approximation. Consider a finite-basis approximation with six basis states

\[
\Psi = \sum_{j=1}^{6} c_j \Phi_j,
\]

where the basis states are

\[
\begin{align*}
\Phi_1 &= |n\rangle|x, 1/2\rangle|s, -1/2\rangle, \\
\Phi_2 &= |n+1\rangle|x, 1/2\rangle|s, -1/2\rangle, \\
\Phi_3 &= |n+1\rangle|x, -1/2\rangle|s, -1/2\rangle, \\
\Phi_4 &= |n+1\rangle|x, 1/2\rangle|s, 1/2\rangle, \\
\Phi_5 &= |n\rangle|x, -1/2\rangle|s, 1/2\rangle.
\end{align*}
\]

The energy levels and coupling are indicated schematically in figure 4. The excitation transfer process in this case would take the system from an initial state \( \Phi_1 \) (with an excited first two-level system, and a ground state second two-level system) to a final state \( \Phi_6 \) (with a ground state first two-level system, and an excited second two-level system).

Since the Hamiltonian \( \hat{H} \) does not couple the basis state \( \Phi_1 \) to the basis state \( \Phi_6 \) directly, the coupling between the two states is indirect. The intermediate states \( \Phi_2, \ldots, \Phi_5 \) will provide the dominant pathways between the initial and final states in the case of weak coupling.

6.2. Indirect coupling

It is possible to obtain an indirect interaction between basis state \( \Phi_1 \) and basis state \( \Phi_6 \) by eliminating the intermediate states algebraically, as we illustrate in what follows. The finite-basis equations for the expansion coefficients \( c_1, \ldots, c_6 \) can be written as

\[
\begin{align*}
Ec_1 &= H_1 c_1 + U_1 \sqrt{n} c_2 + U_1 \sqrt{n+1} c_3 + U_2 \sqrt{n} c_4 + U_2 \sqrt{n+1} c_5, \\
Ec_2 &= H_2 c_2 + U_1 \sqrt{n} c_1 + U_2 \sqrt{n} c_6, \\
Ec_3 &= H_3 c_3 + U_1 \sqrt{n} c_1 + U_2 \sqrt{n} c_6, \\
Ec_4 &= H_4 c_4 + U_2 \sqrt{n} c_1 + U_1 \sqrt{n} c_5, \\
Ec_5 &= H_5 c_5 + U_2 \sqrt{n} c_1 + U_1 \sqrt{n} c_6, \\
Ec_6 &= H_6 c_6 + U_2 \sqrt{n} c_2 + U_2 \sqrt{n+1} c_3 + U_1 \sqrt{n} c_4 + U_1 \sqrt{n+1} c_5.
\end{align*}
\]

The algebraic elimination of the coefficients \( c_2 \) through \( c_5 \) produces two coupled equations of the form

\[
\begin{align*}
Ec_1 &= [H_1 + \Sigma_1(E)] c_1 + V_{16}(E) c_6, \\
Ec_6 &= [H_6 + \Sigma_6(E)] c_6 + V_{61}(E) c_1,
\end{align*}
\]

where

\[
V_{16}(E) = V_{61}(E) = U_1 U_2 \left[ \frac{n}{E - H_2} + \frac{n+1}{E - H_3} + \frac{n}{E - H_4} + \frac{n+1}{E - H_5} \right].
\]
The self-energy terms $\Sigma_1(E)$ and $\Sigma_6(E)$ are small in the case of weak coupling, and contribute to the energy at which the resonance occurs, but not to the splitting.

6.3. Level splitting

Within this model, the resonance condition is

$$H_1 + \Sigma_1(E) = H_0 + \Sigma_6(E)$$

which is the finite-basis approximation equivalent to equation (31) when no oscillator quanta are exchanged in association with the excitation transfer process. The level splitting at resonance in this finite-basis model is

$$\delta E_{\text{min}} = 2\sqrt{V_{16}(E)V_{61}(E)}.$$  (45)

If the self-energy terms can be neglected, then the energy eigenvalue $E$ is very nearly equal to the unperturbed level energy of the initial and final states

$$E = H_1 = H_0.$$  (46)

We can evaluate the indirect coupling term $V_{16}(E)$ at resonance to be

$$V_{16}(E) = \frac{2\hbar\omega_0 U_1 U_2}{(\Delta E_1)^2 - (\hbar\omega_0)^2},$$

where $\Delta E_1$ must approximately the same as $\Delta E_2$ for the resonance condition of equation (46) to be satisfied. When the characteristic oscillator energy $\hbar\omega_0$ is much smaller than the transition energy $\Delta E_1$, then the level splitting on resonance $\delta E_{\text{min}}$ in this approximation evaluates to

$$\delta E_{\text{min}} = \frac{4\hbar\omega_0 g_1 g_2}{n}.$$  (48)

This is equivalent to the splitting we obtained using degenerate perturbation theory in the rotated frame (equation (38)) as long as the integral $I_{12}$ is unity. From an inspection of equation (A.9), we see that this is the case as long as

$$g_1, g_2 \ll 1.$$  (49)

Our result obtained in this section then is the weak coupling limit of what we obtained above. The generalization to a multi-mode system is considered in appendix B.

6.4. Destructive interference and loss

The second-order coupling coefficient $V_{16}(E)$ for indirect coupling between states $\Phi_1$ and $\Phi_6$ is much smaller than the direct coupling coefficients with intermediate states. For example, we may write

$$\frac{V_{16}(E)}{U_1 \sqrt{n}} \approx \frac{2g_2}{n} \frac{\hbar\omega_0}{\Delta E_1}.$$  (50)

This reduction of coupling strength is due to destructive interference between the different contributions in equation (43) that make up $V_{16}(E)$.

To show that this is so, we consider a modified version of the model in which the destructive interference is removed.

Consider the two-spin plus oscillator Hamiltonian augmented with a loss term $-i\hbar\Gamma(E)/2$

$$\hat{H}_1 = \Delta E_1 \frac{\hat{\sigma}_1^{(1)}}{\hbar} + \Delta E_2 \frac{\hat{\sigma}_2^{(2)}}{\hbar} + \hbar\omega_0 \hat{a} \hat{\alpha} - \frac{i\hbar}{2} \hat{\Gamma}(E) + U_1 (\hat{a} \hat{\alpha} + \hat{\alpha} \hat{a}) - \frac{2\hbar\omega_0^{(1)}}{\hbar} + U_2 (\hat{a} \hat{\alpha} + \hat{\alpha} \hat{a}) \frac{2\hbar\omega_0^{(2)}}{\hbar}. $$  (51)

This loss term accounts for energetic decay processes of the low-lying intermediate states at the transition energy of the two-level systems. Only intermediate states with both two-level systems in the ground state ($\Phi_1$ and $\Phi_3$) can decay in this model, since these states have unperturbed energies much less than the available energy $E$. The system cannot decay in such a way as to produce $\Phi_4$ and $\Phi_5$ as final states.

The coefficients $c_2$ and $c_3$ in this kind of model now satisfy

$$E_{c_2} = (H_2 - \frac{\hbar\omega_0}{2}) c_2 + U_1 \sqrt{\rho_1} + U_2 \sqrt{\rho_0},$$

$$E_{c_3} = (H_3 - \frac{\hbar\omega_0}{2}) c_3 + U_1 \sqrt{n + \mathbf{1}} c_1 + U_2 \sqrt{n + \mathbf{1}} c_0.$$  (52)

The indirect coupling coefficient $V_{16}(E)$ in this model is changed due to loss effects. We may write

$$V_{16}(E) = U_1 U_2 \left[ \frac{n}{E - H_2 + i\hbar\Gamma/2} + \frac{n + 1}{E - H_3 + i\hbar\Gamma/2} \right] + \frac{n + 1}{E - H_3 - \hbar\omega_0}.$$  (53)

In the limit that the loss term becomes very large then we may write

$$V_{16}(E) = -\frac{2U_1 U_2 \Delta E_1 n}{(\Delta E_1)^2 - (\hbar\omega_0)^2}(\Gamma \rightarrow \infty).$$  (54)

The ratio of this indirect coupling coefficient (which is now free of destructive interference effects) to the direct coupling coefficient between states $\Phi_1$ and $\Phi_2$ becomes

$$\frac{V_{16}(E)}{U_1 \sqrt{n}} \approx -2g_2.$$  (55)

The removal of destructive interference increases the indirect coupling coefficient by a factor of

$$\frac{|V_{16}(E)|_{\Gamma \rightarrow \infty}}{|V_{16}(E)|_{\Gamma = 0}} = \frac{n \Delta E_1}{\hbar\omega_0}.$$  (56)

In appendix C, we consider a different modification of the model in which the coupling between the different two-level systems and oscillator is through conjugate oscillator operators. Part of the interference is removed in this model.

7. Summary and conclusions

In previous publications, we studied the shifts and splitting of energy levels in the multiphoton regime in the spin-boson model, and in generalizations of the spin-boson model to the case of an oscillator coupled to a spin-one system, and to a three-level system. In these works, we made use of a unitary transformation which rotates the Hamiltonian into a form in which terms primarily responsible for energy level shifts are separated from terms responsible for the level splitting. Here the same general approach is applied to a model involving a pair of two-level systems coupled to an oscillator, where the transition energies and coupling strengths can be different.
The energy levels can be approximated accurately using a WKB approximation based on the unperturbed part of the rotated Hamiltonian \( \hat{H}_0 \), similar to what we found previously. The level splitting at the Bloch–Siegert anticrossings are also described accurately (away from anticrossing resonances) using degenerate perturbation theory based on \( \hat{V}_1 \) and \( \hat{V}_2 \); the situation is very similar to what we found in the spin-boson problem.

What is new in this model (with no analog in the spin-boson problem) is the excitation transfer effect, in which a transition in one two-level system occurs in concert with a transition in the other two-level system. Approximate level energies derived from the unperturbed Hamiltonian \( \hat{H}_0 \) can be used to locate excitation transfer resonances (see figure 2). The excitation transfer effect is mediated by the spin–spin term \( \hat{V}_{12} \) in the rotated Hamiltonian; level splittings away from the Bloch–Siegert (energy exchange) resonances are accurately modeled using degenerate perturbation theory based on the spin–spin term.

It is interesting to consider this effect in the context of a physical system, involving coupling with either spins or electric dipoles in an electromagnetic cavity. If the cavity is very small, it may be that a relatively small number of lower-energy photons will produce a sufficiently strong coupling so that coherent excitation transfer can occur before the onset of decoherence. If one system is initially excited and the other not, then partial excitation transfer will produce entanglement, functionally similar to that proposed in [13, 14] but involving a different kind of coupling. From a conceptual viewpoint, the excitation transfer process under consideration may be counter-intuitive. Excitation of the first system at the dressed transition energy \( \Delta E_1(g_1) \) is transferred to the second system (perhaps with some minor energy exchange with the cavity); however, the cavity does not accept most of the energy which is transferred. Instead, this excitation transfer process, if thought about in perturbation theory, involves an intermediate state consisting of both two-level systems and the cavity which is off resonance. If only a single cavity mode is involved, then this interaction can in principle occur over a larger distance than Coulombic dipole–dipole coupling. The possibility of transferring excitation from an excited source two-level system to another which is resonant (with the exchange of a few photons), located anywhere within a cavity mode, suggests a detection capability.

The effect is weak in this model at large \( n \) due to destructive interference effects. Destructive interference was examined in the context of a finite-basis approximation appropriate to the weak coupling limit of the model, in which contributions from different paths can be seen to cancel. A version of the model augmented with loss in the intermediate states removes the destructive interference, and leads to a drastic increase in the indirect coupling term. A consequence of the destructive interference is that the excitation transfer effect is independent of mode excitation in the weak coupling limit. If there are many modes present, then it is likely that interference between the contributions of the different modes will further decrease the effect, especially if the atoms or molecules represented by the two-level systems are far apart. This effect is discussed in appendix B. Hence, one would expect excitation transfer to be a weak and short range effect in a multi-mode system where the destructive interference involving different pathways is not removed. Such is the case in phonon-mediated excitation transfer.

One question raised from the analysis and discussion presented here concerns the possibility of developing an excitation transfer system in which the destructive interference is reduced or eliminated. In this paper we have noted a reduction of interference in a modified version of the model with conjugate coupling (appendix C), and elimination in a model with large loss in intermediate states (section 6). In either case, the excitation transfer rates and level splitting will be greatly increased. A conjugate coupling scheme could be developed in an electromagnetic resonator under conditions where one two-level system couples to the electric field, and the other two-level system couples to the magnetic field (since electric and magnetic fields in a resonator are conjugate variables). To implement a physical system in which loss impacts the destructive interference, one would require a strongly driven mode which sees low loss at the resonant frequency, but is very lossy at higher frequency corresponding to the transition energies. It will be of interest in future work to make use of these approaches to design new physical systems that are capable of transferring excitation over macroscopic distances.

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Appendix A. WKB approximation

We have found the WKB approximation to be very effective for the large-\( n \) limit of this problem. Within the WKB approximation, the eigenfunctions \( u(y) \) are written in the form [22]

\[
 u(y) = C \frac{\sin \phi}{\sqrt{\eta}},
\]

where

\[
 \frac{d}{dy} \phi(y) = \eta(y) \quad (A.2)
\]

\[
 \eta(y) = \sqrt{\epsilon - v(y)} \quad (A.3)
\]

and where \( C \) is a normalization constant. Written in terms of WKB eigenfunctions, the integral \( I_{12} \) becomes

\[
 I_{12} = C_0 C_1 \int_{-\infty}^{\infty} \frac{\sin[\phi_0(y)] \sin[\phi_1(y)]}{\sqrt{\eta_0(y) \eta_1(y)} (1 + \frac{M_0^2}{\Delta E_1^2}) (1 + \frac{M_1^2}{\Delta E_2^2})} \, dy.
\]

The product of sine functions in the numerator can be decomposed into terms which involve rapid oscillations,

\[
 I_{12} = C_0 C_1 \int_{-\infty}^{\infty} \frac{\sin[\phi_0(y)] \sin[\phi_1(y)]}{\sqrt{\eta_0(y) \eta_1(y)} (1 + \frac{M_0^2}{\Delta E_1^2}) (1 + \frac{M_1^2}{\Delta E_2^2})} \, dy.
\]
Table A1. Results for $I_{12}$ at resonance for $\Delta E_{1} = 11\hbar \omega_{0}$ and $\Delta E_{2} = 15\hbar \omega_{0}$ for $r_{0} = 9600$. Values of the dimensionless coupling constants $g_{1}$ and $g_{2}$ at resonance (first two columns); integral computed from eigenfunctions of $\hat{H}_{0}$ (third column); integral computed from the WKB approximation (fourth column).

| $g_{1}$ | $g_{2}$ | $|I_{12}|$ | $|I_{12}|_{WKB}$ |
|---------|---------|-----------|-----------------|
| 0.50    | 0.161   | 0.2224    | 0.2235          |
| 0.60    | 0.273   | 0.1946    | 0.1951          |
| 0.70    | 0.366   | 0.1733    | 0.1734          |
| 0.80    | 0.452   | 0.1562    | 0.1562          |
| 0.90    | 0.535   | 0.1421    | 0.1420          |
| 1.00    | 0.615   | 0.1303    | 0.1301          |

and slow oscillations. For this, we can make use of the trigonometric identity

$$\sin(\phi_{0}) \sin(\phi_{1}) = \frac{\cos(\phi_{1} - \phi_{0}) - \cos(\phi_{1} + \phi_{0})}{2}.$$  \hspace{1cm} (A.5)

On the rhs, the first cosine function has a slowly varying phase, and the second one has a rapidly varying phase (especially if $n$ is very large). Usually the contribution from the cosine with the slowly varying phase dominates, in which case we may write

$$I_{12} = \frac{C_{0}C_{1}}{2} \int_{-\infty}^{\infty} \cos[\Delta \phi(y)] \eta(y) \left(1 + \frac{8l_{1}^{2}y^{2}}{\Delta E_{1}^{2}}\right) \left(1 + \frac{8l_{2}^{2}y^{2}}{\Delta E_{2}^{2}}\right) \, dy.$$ \hspace{1cm} (A.6)

In writing this we assume that the two WKB momentum variables $\eta_{0}$ and $\eta_{1}$ are not very different

$$\eta_{0}(y) \approx \eta_{1}(y) \rightarrow \eta(y).$$ \hspace{1cm} (A.7)

The normalization constants $C_{0}$ and $C_{1}$ in this approximation satisfy

$$C_{0}^{2} \int_{-\infty}^{\infty} \frac{1}{\eta(y)} \, dy = 1.$$ \hspace{1cm} (A.8)

This allows us to recast the WKB approximation in the form

$$I_{12} = \frac{1}{\eta(y)} \int_{-\infty}^{\infty} \cos[\Delta \phi(y)] \eta(y) \left(1 + \frac{8l_{1}^{2}y^{2}}{\Delta E_{1}^{2}}\right) \left(1 + \frac{8l_{2}^{2}y^{2}}{\Delta E_{2}^{2}}\right) \, dy \int_{-\infty}^{\infty} \frac{1}{\eta(y)} \, dy.$$ \hspace{1cm} (A.9)

In table A1 we give the results of computations of the magnitude of the integral $|I_{12}|$ from the numerical solution of equation (17), and from the WKB approximation of equation (A.9). One sees that the WKB approximation in this case is very close to the numerically exact result.

**Appendix B. Multi-mode case**

There is an additional mechanism that produces destructive interference in the multi-mode generalization of this problem. In particular, interference can occur between the contributions of the different modes.

**B.1. Multi-mode Hamiltonian**

For simplicity, suppose that we have many modes that can be described in terms of wavevectors $k$; and that the mode frequencies are a function of the wavevectors according to the dispersion relation

$$\omega = \omega(k) \rightarrow \omega_{k}.$$ \hspace{1cm} (B.1)

The multi-mode generalization of the two-spin problem then might be described according to a Hamiltonian $\hat{H}_{mm}$ of the form

$$\hat{H}_{mm} = \Delta E_{1} \frac{\partial^{2}}{\partial \phi_{1}^{2}} + \Delta E_{2} \frac{\partial^{2}}{\partial \phi_{2}^{2}} + \sum_{k} \hbar \omega_{k} \hat{a}_{k}^{\dagger} \hat{a}_{k}$$

$$+ \sum_{k} \left[U_{1}(k)\hat{a}_{k}^{\dagger} e^{ikr_{1}} + U_{1}^{*}(k)\hat{a}_{k} e^{-ikr_{1}}\right] \frac{2\Phi_{1}(k)}{\hbar}$$

$$+ \sum_{k} \left[U_{2}(k)\hat{a}_{k}^{\dagger} e^{ikr_{2}} + U_{2}^{*}(k)\hat{a}_{k} e^{-ikr_{2}}\right] \frac{2\Phi_{2}(k)}{\hbar},$$ \hspace{1cm} (B.2)

where $r_{1}$ and $r_{2}$ are the position vectors of the two atoms.

**B.2. Indirect coupling without loss**

It is possible to develop a finite-basis model similar to that discussed in section 6, but now for the multi-mode case, to obtain the indirect coupling term for the weak-coupling limit. If we carry out such a calculation, in place of equation (47), we obtain

$$V_{16}(E) = \frac{2\hbar \omega_{h} U_{1} U_{2}}{(\Delta E_{1})^{2} - (\hbar \omega_{k})^{2}} \int \frac{U_{1}(k)U_{1}^{*}(k) e^{ik(r_{1} - r_{2})}}{\Delta E_{1} - \hbar \omega_{k}} \, d^{3}k$$

$$- \int \frac{U_{2}^{*}(k)U_{2}(k) e^{-ik(r_{1} - r_{2})}}{\Delta E_{1} + \hbar \omega_{k}} \, d^{3}k.$$ \hspace{1cm} (B.3)

The contribution of a single highly off-resonant mode appears on the same footing as contributions from many other off-resonant modes, each weighted by a phase factor. In the event that the separation $|r_{1} - r_{2}|$ is large, then severe cancellation can occur between the contributions of the different modes; hence we would expect interactions to be of short range in the multi-mode case. Indirect coupling through coupling to common phonon modes would be described using such an approach (see for example [23]). If one adopts for the multi-mode oscillator longitudinal photon modes (all with zero energy), then indirect coupling between electric dipoles can be thought of in this way; with an overall $|r_{1} - r_{2}|^{-3}$ dependence of the dipole–dipole matrix element in the absence of retardation for single longitudinal photon exchange. Excitation transfer in this case is known as resonance energy transfer in biophysics [18].

In general, any long range interactions in this kind of model will be dominated by resonant interactions if resonant modes exist. Otherwise, the indirect interaction will most likely occur only at short range.

**B.3. Indirect coupling with loss**

If some mechanism is present that can remove the destructive interference effect in the different pathways, then it may be possible to reduce or eliminate the destructive interference...
effect in the multi-mode version of the problem. The Hamiltonian in this case can be written as

\[ \hat{H}_{\text{multi}} = \Delta E_1 \frac{\hat{a}^{\dagger}}{\hbar} + \Delta E_2 \frac{\hat{a}}{\hbar} + \sum_k \hbar \omega_k \hat{a}_k^{\dagger} \hat{a}_k^{\dagger} - \frac{i \hbar \Gamma(E)}{2} \]

\[ + \sum_k \left[ U_1(k) \hat{a}_k e^{ik \cdot r_1} + U_1^\dagger(k) \hat{a}_k e^{-ik \cdot r_1} \right] \frac{2 \hat{a}^{\dagger}}{\hbar} \]

\[ + \sum_k \left[ U_2(k) \hat{a}_k e^{ik \cdot r_2} + U_2^\dagger(k) \hat{a}_k e^{-ik \cdot r_2} \right] \frac{2 \hat{a}^{\dagger}}{\hbar} \quad \text{(B.4)} \]

An analogous finite-basis type model can be developed for this kind of model, leading to an indirect coupling in the \( \Gamma \to \infty \) limit of

\[ V_{16}(E) = \frac{2 U_1 U_2 \Delta E_1 \hbar}{(\Delta E_1)^2 - (\hbar \omega_0)^2} \]

\[ - \int \frac{U_1(k) U_2(k) n(k) e^{i k \cdot (r_1 - r_2)}}{\Delta E_1 - \hbar \omega_k} d^3k \]

\[ - \int \frac{U_1^\dagger(k) U_2^\dagger(k) [n(k) + 1] e^{-i k \cdot (r_1 - r_2)}}{\Delta E_1 + \hbar \omega_k} d^3k. \quad \text{(B.5)} \]

Things are qualitatively different in this case, since the indirect coupling now depends on the number of quanta \( n(k) \) in the different modes. If a single off-resonant mode is very highly excited, then the interaction can take place over a much longer range since the unexcited mode can no longer destructively interfere.

**Appendix C. Model with conjugate couplings**

It is possible to eliminate some of the destructive interference by allowing the two different two-level systems to couple with the oscillator through interactions that do not commute. Consider a modified version of the model described by the Hamiltonian

\[ \hat{H}_c = \Delta E_1 \frac{\hat{a}^{\dagger}}{\hbar} + \Delta E_2 \frac{\hat{a}}{\hbar} + \hbar \omega_0 \hat{a} + U_1 (\hat{a}^{\dagger} + \hat{a}) \frac{2 \hat{a}^{\dagger}}{\hbar} \]

\[ + i U_2 (\hat{a}^{\dagger} - \hat{a}) \frac{2 \hat{a}^{\dagger}}{\hbar}. \quad \text{(C.1)} \]

If we adopt the same finite-basis approximation for the wavefunction \( \Psi \) as discussed in section 6

\[ \Psi = \sum_{j=1}^{6} c_j \Phi_j \quad \text{(C.2)} \]

then the expansion coefficients satisfy

\[ E_{c_1} = H_c C_1 + U_1 \sqrt{n_c} \hat{c}_3 + U_1 \sqrt{n + 1} \hat{c}_3 + i U_2 \sqrt{n} \hat{c}_4 \]

\[ - i U_2 \sqrt{n + 1} \hat{c}_4 \]

\[ E_{c_2} = H_c C_2 + U_1 \sqrt{n_c} \hat{c}_3 + i U_2 \sqrt{n} \hat{c}_6, \]

\[ E_{c_3} = H_c C_3 + U_1 \sqrt{n + 1} \hat{c}_3 - i U_2 \sqrt{n + 1} \hat{c}_6, \]

\[ E_{c_4} = H_c C_4 - i U_2 \sqrt{n} \hat{c}_6 + U_1 \sqrt{n} \hat{c}_6, \]

\[ E_{c_5} = H_c C_5 + i U_2 \sqrt{n + 1} \hat{c}_3 + U_1 \sqrt{n + 1} \hat{c}_6, \]

\[ E_{c_6} = H_c C_6 + i U_2 \sqrt{n_c} \hat{c}_2 - i U_2 \sqrt{n + 1} \hat{c}_3 + U_1 \sqrt{n} \hat{c}_4 \]

\[ + U_1 \sqrt{n + 1} \hat{c}_4. \quad \text{(C.3)} \]

The indirect coupling coefficient \( V_{16}(E) \) in this case is

\[ V_{16}(E) = i U_1 U_2 \left[ \frac{n}{E - H_3} - \frac{n + 1}{E - H_3} + \frac{n}{E - H_4} - \frac{n + 1}{E - H_5} \right]. \quad \text{(C.4)} \]

At resonance, we assume that

\[ E = H_1 = H_6 \quad \text{(C.5)} \]

to obtain

\[ V_{16}(E) = - \frac{2 i U_1 U_2 \hbar \omega_0 (2n + 1)}{(\Delta E_1)^2 - (\hbar \omega_0)^2}. \quad \text{(C.6)} \]

This indirect coupling is much larger than that we obtained with the original model, and also depends on the mode excitation. However, we see that it is smaller in magnitude by \( \hbar \omega_0 / \Delta E_1 \) than the result (equation (54)) we obtained in which the destructive interference was completely eliminated by loss. Hence, we have removed some of the destructive interference with conjugate coupling.

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