Multiphoton Bloch–Siegert shifts and level-splittings in spin-one systems

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Abstract
We consider a spin-boson model in which a spin-1 system is coupled to an oscillator. A unitary transformation is applied which allows a separation of terms responsible for the Bloch–Siegert shift, and terms responsible for the level splittings at anticrossings associated with Bloch–Siegert resonances. When the oscillator is highly excited, the system can maintain resonance for sequential multiphoton transitions. At lower levels of excitation, resonance cannot be maintained because energy exchange with the oscillator changes the level shift. An estimate for the critical excitation level of the oscillator is developed.

1. Introduction
The interaction of a two-level system with oscillatory off-diagonal coupling leads to a shift in the transition energy, known in the literature as the Bloch–Siegert shift [1, 2]. When the shifted transition energy is resonant with an odd number of oscillator quanta, energy exchange between the two systems can occur. This effect appears in energy level calculations as a level splitting at the Bloch–Siegert resonances. Both the shift and splitting have been studied previously using models based on the Rabi Hamiltonian (in which the oscillator is presumed classical) [1–6] and based on the spin-boson Hamiltonian (in which the oscillator is presumed quantum mechanical) [7, 8]. It has been noted that the Bloch–Siegert shift arises from the counter-rotating terms in the Hamiltonian, and that it disappears when the rotating-wave approximation is made [8, 9].

Our interest in this problem is motivated by the possibility of coherent energy exchange between quantum systems with highly mismatched characteristic energies. For example, we have been interested in the dynamics of energy exchange between a two-level system with a large transition energy $\Delta E$, and an oscillator with a small energy quantum $\hbar\omega$. The spin-boson Hamiltonian is one of the simplest models exhibiting such coherent energy exchange. We recently considered the Bloch–Siegert shift [10] and level splittings [11] at the Bloch–Siegert resonances, the latter of which is a result of the coherent energy exchange between the two-level system and oscillator.

Most of the work cited above is focused on the two-level problem, which is usually modelled as a spin-1/2 system interacting with a single harmonic oscillator. The generalization of the problem to systems involving higher spin leads to more complicated models which have not received comparable attention in the literature. The Bloch–Siegert shift for the spin-1 case was studied previously by Hermann and Swain [12, 13]. Some progress has also been made in the case of the general spin problem [8].

In this work, we focus on the problem of a spin-1 system coupled to a simple harmonic oscillator. Our earlier analysis in the spin-1/2 version of the problem made use of a unitary transform in which the shift and level splitting effects can be associated separately with different terms in the rotated Hamiltonian. Such a separation serves as the basis for the development of analytic results for both shift and splitting which are useful over a wider range of coupling strength than that available previously. The unitary transform that we used for the spin-1/2 problem can be extended simply to higher spin models, allowing for a separation of shift and level splitting effects in more complicated problems. We have decided to focus here on the spin-1 case since it provides a good example of this generalization.

Although the spin-1 system is a three-state system, we have found it useful to think about it in terms of an underlying two-spin problem. For example, the Bloch–Siegert shift that we find below for the spin 1 can be understood simply as arising from the individual shifts associated with two spin-1/2 systems. Later on in this work, we find that the Bloch–Siegert resonance condition cannot be maintained at modest $n$, which can be understood in terms of an initial resonant spin-1/2 transition that exchanges energy with the oscillator, followed
by a second spin-1/2 transition that is no longer resonant since the oscillator is changed.

2. Unitary equivalent Hamiltonian

We focus on a spin-1 system coupled to an oscillator, using a spin-boson Hamiltonian of the form

\[
\hat{H} = \Delta E \frac{\hat{S}_z}{\hbar} + \hbar \omega \hat{a}^\dagger \hat{a} + U (\hat{a}^\dagger + \hat{a}) 2 \frac{\hat{S}_x}{\hbar}.
\]

(1)

In the above equation, \( U \) represents the interaction between the harmonic oscillator and the spin system. We are interested in the regime where the photon excitation \( n_0 \) is large, and where the transition energy is much greater than an oscillator quantum (\( \Delta E \gg \hbar \omega \)).

As was the case in the spin-1 problem [10], it is useful to consider in the case of the spin-1 problem the unitary equivalent Hamiltonian

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

(2)

where

\[
\hat{U} = \exp \left\{ -\frac{i}{2} \arctan \left[ \frac{2U (\hat{a}^\dagger + \hat{a})}{\Delta E} \right] \right\}
\]

The rotated Hamiltonian becomes

\[
\hat{H}' = \hat{H}_0 + \hat{V} + \hat{W}
\]

(3)

where

\[
\hat{H}_0 = \sqrt{\Delta E^2 + 4U^2 (\hat{a}^\dagger + \hat{a})^2} \frac{\hat{S}_z}{\hbar} + \hbar \omega \hat{a}^\dagger \hat{a}
\]

(4)

\[
\hat{V} = \frac{\hbar \omega_0}{2} \left\{ \left[ \frac{U}{\Delta E} \right] (\hat{a} - \hat{a}^\dagger) + (\hat{a} - \hat{a}^\dagger) \left[ \frac{U}{\Delta E} \right] \right\} \frac{2 \hat{S}_x}{\hbar}
\]

(5)

\[
\hat{W} = \hbar \omega_0 \left\{ \frac{U}{\Delta E} \frac{2 \hat{S}_x}{\hbar} \right\}^2
\]

(6)

Within the parameter space of interest to us, the \( \hat{W} \) term is small; consequently, we neglect it in what follows.

3. Approximate energy eigenvalues

In the large-\( n \) limit, the oscillator has a strong impact on the spin system, but the spin system impacts the oscillator only weakly. As a result, the energy levels are found to be reasonably well approximated away from the level anticrossings by

\[
E_{n,M}(\omega) = \Delta E (\omega) M + \hbar \omega_0 n
\]

(7)

where \( \Delta E (\omega) \) is the dressed two-level system energy. This is very much like the behaviour found for the spin-1/2 version of the problem discussed previously [10]. It is useful to adopt the same definition for the dimensionless coupling constant \( g \) for the spin-1 version of the problem

\[
g = \frac{U \sqrt{\omega_0}}{\Delta E}
\]

that we used before for the spin-1/2 case.

The dressed two-level system energy \( \Delta E (g) \) is also the same, as would be expected since it comes about from the same basic interaction. We can think of the spin-1 system as being made up of two spin-1/2 systems coupled identically to the same oscillator. Since the Bloch–Siegert shift is the result of the interaction between a single two-level system and oscillator, we should expect that it will be nearly the same per two-level system if there are more than one.

We can make use of the rotation to see this. Consider eigenfunctions of the unperturbed \( \hat{H}_0 \) part of the rotated Hamiltonian

\[
E_{n,M} \psi_{n,M} = \hat{H}_0 \psi_{n,M}
\]

(8)

We can separate variables (as done in the spin-1/2 case) to write

\[
\psi_{n,M} = |S, M\rangle u_{n,M}(y)
\]

where \( u_{n,M}(y) \) satisfies

\[
\left(E_{n,M} + \frac{\hbar \omega_0}{2}\right) u_{n,M} = \frac{\hbar \omega_0}{2} \left[ \frac{d^2}{dy^2} + y^2 \right] u_{n,M} + M \sqrt{\Delta E^2 + 8U^2(\hat{a}^\dagger + \hat{a})^2} u_{n,M}.
\]

(9)

Here, we use spin-1 eigenfunctions \( |S, M\rangle \), with \( S = 1 \) and \( M = 0, \pm 1 \). In the large-\( n \) limit, this is just a modified simple harmonic oscillator with a small \( M \)-dependent perturbation. By adopting a simple harmonic oscillator wavefunction as a trial solution in a variational computation, we obtain approximate energy levels of the form of equation (7) where

\[
\Delta E (g) = \langle n | \sqrt{\Delta E^2 + 8U^2(\hat{a}^\dagger + \hat{a})^2} | n \rangle.
\]

(10)

This dressed transition energy, identical to what we found in the spin-1/2 case [10], is in good agreement with the numerical results away from the level anticrossings.

4. Multiphoton resonances

As mentioned above, our interest in this model is driven in part by the possibility of coherent energy exchange between systems with strongly mismatched characteristic energies. In the spin-1 problem, resonances occur associated with anticrossings when the dressed two-level system energy \( \Delta E (g) \) matches an odd number of oscillator quanta. In the large-\( n \) limit, the oscillator is impacted only weakly through the exchange of a modest number of oscillator quanta; in which case the dressed energy \( \Delta E (g) \) is approximately invariant, and we are able to develop a resonance condition applicable to both transitions. When \( n \) is smaller, the change in the number of oscillator quanta may be a significant fraction of the total number of oscillator quanta; in which case the dressed two-level transition energy may be on resonance for one transition and off resonance for the other. We begin our discussion here focusing on the first case, since it is simpler.
4.1. Large-n resonance condition

We are interested then in the resonance condition at which the dressed transition energy is matched to an odd number of oscillator quanta. To proceed, we consider three basis states which are eigenstates of the rotated Hamiltonian $\hat{H}_0$:

\[
\begin{align*}
\phi_{-1} &= u_{n+\Delta n,-1}(y)|S,-1\rangle \\
\phi_0 &= u_{n,0}(y)|S,0\rangle \\
\phi_1 &= u_{n-\Delta n,1}(y)|S,1\rangle
\end{align*}
\]

(11)

where $S = 1$. These states have energies $\epsilon_{-1}$, $\epsilon_0$ and $\epsilon_1$ respectively, which in the large-$n$ limit are given approximately by

\[
\begin{align*}
\epsilon_{-1} &= \hbar \omega_0(n + \Delta n) - \Delta E(g) \\
\epsilon_0 &= \hbar \omega_0 n \\
\epsilon_1 &= \hbar \omega_0(n - \Delta n) + \Delta E(g)
\end{align*}
\]

(12-14)

with $\Delta E(g)$ taken to be a constant for both transitions. The resonance condition in this case is

\[\Delta E(g) = \Delta n\hbar \omega_0\]

(15)

where, as in the two-level case, the number of oscillator quanta exchanged $\Delta n$ must be odd for level splitting to occur.

4.2. Comparison with previous work

As mentioned above, Hermann and Swain [13] have calculated the Bloch–Siegert shift for the spin-1 case. In their notation, the resonance condition to fourth order is

\[
\omega_{\min} = \frac{\omega_0}{p} + \frac{2V^2p}{(p^2 - 1)\omega_0} - \frac{p^3(3p^2 - 7)V^4}{(p^2 - 1)^3\omega_0^3}.
\]

(16)

In order to write this resonance condition in our notation, we make the following replacements

\[
V \rightarrow U\sqrt{2n}, \quad \omega_{\min} \rightarrow \omega_0, \quad \omega_0 \rightarrow \Delta E, \quad p \rightarrow (2k + 1)
\]

to obtain

\[
\Delta E \left[1 + \frac{4g^2(2k + 1)^2}{4k(k + 1)} - \frac{4(2k + 1)^4[3(2k + 1)^2 - 7]g^4}{[4k(k + 1)]^3} + \cdots \right] = (2k + 1)\hbar \omega_0,
\]

(17)

Note that equation (17) is exactly the same condition as obtained by Ahmad and Bullough [3, 10] for the spin-1/2 problem.

To compare our calculation to these perturbative results, we can expand $\Delta E(g)$ from equation (10) in powers of $g$ to obtain the condition for the $(2k + 1)$th resonance as

\[
\Delta E(1 + 4g^2 - 12g^4 \cdots) = (2k + 1)\hbar \omega_0
\]

(18)

where $\Delta n = 2k + 1$. This condition is the same as was found in the spin-1/2 problem [10]. In the limit that $k \gg 1$, we see that equation (17) reduces to our result (equation (18)).

5. Dynamics: resonant case

The basis states that we chose (equation (11)) are matched total energy states; the spin-1 system pieces are combined with modified oscillator states in which an odd number of oscillator quanta are matched to the dressed two-level transition energy. When the coupled system makes transitions between these different basis states, the resulting dynamics describe coherent energy exchange between component quantum systems with very different characteristic energies. The resonant case in the high-$n$ limit is most interesting in this regard, since both two-level systems make transitions, and a full $2\Delta n$ oscillator quanta are exchanged.

5.1. A three-state model

To proceed, we consider a dynamical state $\psi(t)$ constructed from the three basis states listed in equation (11):

\[
\psi(t) = c_{-1}(t)\phi_{-1} + c_0(t)\phi_0 + c_1(t)\phi_1.
\]

(19)

In the large-$n$ limit, these three basis states become degenerate. In addition, the different matrix elements become roughly equal

\[
\langle \phi_{-1}|\hat{V}|\phi_0 \rangle \approx \langle \phi_0|\hat{V}|\phi_1 \rangle \rightarrow v
\]

(20)

where

\[
v = \left[\frac{2\hbar \alpha U}{\Delta E}\right] I
\]

(21)

and

\[
\int_{-\infty}^{\infty} u_{n+\Delta n,M}(y) \frac{1}{1 + 8(\frac{\Delta E}{\hbar \omega_0})^2} \left(\frac{d}{dy}u_{n,M}(y)\right) dy \rightarrow I
\]

(22)

\[
-\int_{-\infty}^{\infty} \left(\frac{d}{dy}u_{n+\Delta n,M}(y)\right) \frac{1}{1 + 8(\frac{\Delta E}{\hbar \omega_0})^2} u_{n,M}(y) dy \rightarrow I.
\]

(23)

The two integrals appearing here have been found to approach a common limit (to within the sign) based on calculations using numerical solutions for the rotated frame $\hat{H}_0$ problem, and based on calculations using the WKB approximation. This behaviour can be understood simply by noting that the derivative can be expressed in terms of raising and lowering operators and in the large-$n$ limit the $u_{\pm}(y)$ functions are nearly pure harmonic oscillator states.

The inclusion of the perturbation $\hat{V}$ leads to the restricted Schrödinger equation

\[
\frac{d}{dt} \begin{pmatrix}
\begin{bmatrix} c_{-1} \\ c_0 \\ c_1 \end{bmatrix}
\end{pmatrix} = \begin{pmatrix}
\epsilon_{-1} & 0 & 0 \\
0 & \epsilon_0 & 0 \\
0 & 0 & \epsilon_1
\end{pmatrix} \begin{pmatrix}
\begin{bmatrix} c_{-1} \\ c_0 \\ c_1 \end{bmatrix}
\end{pmatrix}.
\]

(24)

5.2. Dynamical solution

Solutions for equation (24) can be readily constructed using an eigenfunction expansion. One interesting solution is

\[
\begin{pmatrix}
\begin{bmatrix} c_{-1}(t) \\ c_0(t) \\ c_1(t) \end{bmatrix}
\end{pmatrix} = e^{-i\sigma\phi/\hbar} \begin{pmatrix}
\begin{bmatrix} 1 \\ \sqrt{2} \sin \frac{\sigma\phi}{\hbar} \\ \frac{1}{2} \cos \frac{\sigma\phi}{\hbar} + 1
\end{bmatrix} \end{pmatrix}.
\]

(25)
One sees in this case a complete energy exchange between the two systems, where the excitation energy of two dressed two-level systems \(2\Delta E(g)\) is exchanged for an equal amount of oscillator energy \(2\Delta n\hbar\omega_0\).

5.4. Oscillations of \(\langle M \rangle\) are sinusoidal

We can look at the dynamics of energy exchange in the large-\(n\) resonance case in another way. Within the set of basis states that we have selected, the basis states are degenerate on resonance, and the coupling is proportional to matrix elements of \(\hat{S}_z\) (see equation (24)). In this case, we can determine the evolution of \(\langle \hat{S}_z \rangle\) for the spin-1 case (or in other cases as well) through commutation with the interaction term of the associated restricted Hamiltonian. The result is

\[
\frac{d}{dt}\langle \hat{S}_z \rangle = -\Omega^2 \langle \hat{S}_z \rangle
\]

where the frequency \(\Omega\) is

\[
\Omega = \sqrt{2\omega_0}U/\Delta E.
\]

Hence if all the two-level systems are initially in the ground state (in the rotated frame), in general the average \(\langle \hat{S}_z \rangle\) will exhibit sinusoidal dynamics according to

\[
\langle \hat{S}_z \rangle = \langle M \rangle = -S \cos(\Omega t).
\]

This is consistent with the results shown in figure 2. Note that these dynamics are in the rotated frame, and could be obtained if the interaction \(U\) were turned on adiabatically.

6. Finite-\(n\) effects

Up to this point, we have assumed that \(n\) is ‘large enough’ so that the basis states are degenerate. However, when \(n\) is not overly large, the levels are not degenerate and hence it is not possible to arrange for a clean energy exchange as described above. This motivates an interest in understanding how large \(n\) must be so that the system acts as if it is degenerate.

6.1. Parameterization of basis state energies

For finite \(n\), we have found (based on numerical calculations) that the energy levels of the original Hamiltonian \(\hat{H}\) can be accurately fit away from resonance using the expansion of the form

\[
E_{n,M} = A + B(n - n_0) + CM + DM(n - n_0) + FM^2 + \cdots
\]

(30)

where \(A, B, C, D, E\) and \(F\) are fitting coefficients. The terms quadratic in \(\Delta n\) are not that significant because the oscillator is highly excited. We can relate the basis state energies \(\epsilon_{-1}\) and \(\epsilon_1\) to \(\epsilon_0\) using this parameterization to obtain

\[
\epsilon_{-1} = \epsilon_0 + B \Delta n - C - D \Delta n + F + \cdots
\]

(31)

\[
\epsilon_1 = \epsilon_0 - B \Delta n + C - D \Delta n + F + \cdots.
\]

(32)

We take the approximate resonance condition to be

\[
B \Delta n = C
\]

(33)

which is nearly equivalent to our resonance condition from above

\[
\Delta n\hbar\omega_0 = \Delta E(g).
\]

(34)
6.2. Mismatch in basis state energies at resonance

When this resonance condition is satisfied, the energy levels (in the absence of coupling terms) are still not matched. Instead, we find that \( \epsilon_0 \) lies above \( \epsilon_1 \) and \( \epsilon_{-1} \) due to the presence of higher-order terms in the fitting expansion

\[
\epsilon_{-1} = \epsilon_0 - D\Delta n + F + \cdots \quad (35)
\]

\[
\epsilon_1 = \epsilon_0 - D\Delta n + F + \cdots \quad (36)
\]

Both analytic and numeric computations lead to the conclusion that

\[
F - D\Delta n \rightarrow \text{constant} \quad (37)
\]

for large \( n_0 \). The numerator on the RHS is a constant that can be determined from parameterizing the levels in a direct numerical calculation, or can be estimated from perturbation theory in the rotated frame. For the purposes of discussion, we may write this as

\[
F - D\Delta n \rightarrow \left[ n_0(F - D\Delta n) \right]_{n_0=\infty} \quad (38)
\]

6.3. Determination of critical \( n \) where splitting matches coupling

The matrix element that produces transitions in the rotated frame can be approximated by

\[
\langle \phi_0 | \hat{V} | \phi_1 \rangle = \frac{2\hbar\omega_0 U}{\Delta E} I \rightarrow 2\hbar\omega_0 g \left[ \frac{I}{\sqrt{n_0}} \right]_{n_0=\infty} \quad (39)
\]

We can now estimate the number of oscillator quanta \( n \) required to make the coupling matrix element equal in magnitude to the basis state splitting by requiring

\[
|\epsilon_1 - \epsilon_0| = \left[ \frac{n_0(F - D\Delta n)}{n_0} \right]_{n_0=\infty} = \sqrt{2} |\langle \phi_0 | \hat{V} | \phi_1 \rangle| \quad (40)
\]

The \( \sqrt{2} \) that appears here reflects the extra factor in the level splittings obtained from a diagonalization of the three-state model on resonance. This is satisfied when \( n \) is equal to a critical value

\[
n_{\text{crit}} = \left| \frac{n_0(F - D\Delta n)}{2\sqrt{2}\hbar\omega_0 g[I/\sqrt{n_0}]_{n_0=\infty}} \right| \quad (41)
\]

6.4. Numerical example

In figure 3, is shown the level splittings for state computed from the original Hamiltonian \( \hat{H} \) without rotation in an example that illustrates this effect. In this calculation, we have selected a model with \( \Delta E = 11\hbar\omega_0 \), and we focus on the resonance at \( \Delta E(g) = 15\hbar\omega_0 \), where \( g \) is determined from numerical optimization to minimize the level splitting at each \( n_0 \). The numerical data show a resonance behaviour similar to what we would expect from the three-state model above. At large \( n_0 \), the basis states are nearly degenerate relative to the coupling matrix element, and we find maximum level splitting. At small \( n_0 \), the basis states are separated by more than the coupling matrix element, so that the second-order coupling between \( \phi_{-1} \) and \( \phi_1 \) is small, and hence the level splitting is also small. The critical number of oscillator quanta in this case is about \( 2.7 \times 10^4 \).

Figure 3. Level splitting from direct solution of original Hamiltonian \( \hat{H} \) problem as a function of \( n_0 \). The level splitting is between states that in the rotated frame are mostly composed of \( M = -1, \Delta n = 15 \) and \( M = 1, \Delta n = -15 \), for a model in which \( \Delta E = 11\hbar\omega_0 \).

6.5. Estimation of \( n_{\text{crit}} \) from analytic estimate

For this problem, the quantity \( I/\sqrt{n} \) is found from a WKB calculation to be

\[
\left[ \frac{I}{\sqrt{n_0}} \right]_{n_0=\infty} = 1.77 \times 10^{-3} \quad (42)
\]

The \( D \) term estimated from

\[
D = \left( \frac{\partial^2}{\partial n \partial M} E_{n,M} \right)_{n=n_0, M=0} \quad (43)
\]

is used to obtain

\[
n_0 D \rightarrow 6.60 \left( \frac{1}{2\hbar\omega_0} \right) \quad (44)
\]

as \( n_0 \) goes to \( \infty \). From a direct parameterization of the energy levels we obtain

\[
n_0 F \rightarrow -10.4 \left( \frac{1}{2\hbar\omega_0} \right) \quad (45)
\]

We can compare this result from the value obtained using the WKB approximation

\[
n_0 F = \frac{n_0}{2} \left( \frac{\partial^2}{\partial M^2} E_{n,M} \right) = -11.0 \left( \frac{1}{2\hbar\omega_0} \right) \quad (46)
\]

These parameters are combined to produce the estimate based on equation (41)

\[
n_{\text{crit}} = 3.26 \times 10^4 \quad (47)
\]

This estimate is in reasonable agreement with the calculation of figure 3. We conclude that a simple three-state model provides a good foundation for estimating how large \( n \) must be in order for the basis states to be degenerate relative to the coupling.
7. Discussion

We have previously applied a rotation in the case of the spin-1/2 problem in order to produce a dressed system in which the unperturbed Hamiltonian \( \hat{H}_0 \) provides a good approximation away from the level anticrossings at resonance [10], and where the perturbation \( \hat{V} \) gives most of the coupling responsible for the level splittings at the anticrossings [11]. Here we have applied a similar rotation to the spin-1 problem, in which case a similar separation of the rotated Hamiltonian occurs. Although our discussion here is focused on the spin-1 problem, we have found that similar good results are also obtained in the case of higher spin as well. We find that the Bloch–Siegert shift in the spin-1 case is close to that of the spin-1/2 problem, in agreement with the perturbative result of Hermann and Swain [13].

We have considered level splitting at the anticrossing in association with the system dynamics at resonance. In the large-\( n \) limit, energy exchange between the spin-1 system and oscillator constitutes only a small perturbation to the oscillator, so that the dressed transition energy is not changed, and that the same resonance condition applies to both transitions. In this case, it is possible for complete energy exchange to occur between the spin-1 system and the oscillator. We have given an analytic solution for the dynamics in the case of resonance.

At more modest \( n \), the oscillator is modified sufficiently by a change of \( \Delta n \) oscillator quanta so that the resonance condition no longer holds for a second transition. In this case, we might think of the coupled system as being made up of two spin-1/2 systems with different dressed transition energies weakly coupled to the oscillator. Accordingly, at larger \( n \) we might think of the coupled system as a dressed spin-1 system weakly coupled to the oscillator. The transition between these two kinds of behaviour is determined by a critical excitation of the oscillator. We have developed an analytic estimate for this critical number of oscillator quanta \( n_{\text{crit}} \) (see equation (41)) in terms of fitting parameters which we can derive directly from solutions of the unrotated Hamiltonian \( \hat{H} \), from numerical solutions of the unperturbed part of the rotated Hamiltonian \( \hat{H}_0 \), or from the WKB approximation as applied to the rotated problem.

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