J. Condensed Matter Nucl. Sci. 11 (2013) 59–92

Research Article

Lossy Spin–boson Model with an Unstable Upper State and Extension to N-level Systems

Peter L. Hagelstein *
Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

Irfan U. Chaudhary
Department of Computer Science and Engineering, University of Engineering and Technology. Lahore, Pakistan

Abstract

In the Fleischmann–Pons experiment, excess heat is thought to have a nuclear origin due to the amount of energy produced, yet there are no commensurate energetic particles. This has motivated us over the years to focus attention on models in which a large quantum is fractionated into a great many small quanta. We have found that many two-level systems with a large transition energy are able to exchange energy coherently with an oscillator with a much smaller characteristic energy as long as decay channels are present in the vicinity of the two-level transition energy. In previous work we analyzed this basic model, and obtained estimates for the coherent energy exchange rate in the strong coupling limit. In this work we consider a version of this model where the upper states of the two-level systems are unstable. In this case, there is no coherent energy exchange, but instead we find a dynamical polarization effect which we have analyzed. We extend the model to the case of three-level systems, and generalize the result to apply to general N-level systems. Coherent energy exchange is possible within the context of a donor and receiver model, where the receiver transitions have unstable upper states. We give results for the donor dynamics in this case. This model provides a foundation for a new kind of model that we put forth recently for which the predictions appear to be closely connected to experiment.

Keywords: Coherent energy exchange, Fleischmann–Pons experiment, Lossy spin–boson model, Theory

1. Introduction

For the past several years we have pursued models involving sets of two-level systems and a highly excited oscillator with linear coupling and loss [1–4]. These models are interesting to us because they show coherent energy exchange between the two different quantum systems under conditions where the two-level transition energy is much greater than the characteristic energy of the oscillator; in essence, the large quantum is "fractionated" in these models [4].

*E-mail: plh@mit.edu

© 2013 ISCMNS. All rights reserved. ISSN 2227-3123

Our motivation for these studies has be to understand physical mechanisms associated with excess heat production in the Fleischmann–Pons experiment [5–7]. What makes the experimental results difficult to understand is that a large amount of energy is released which appears to be of nuclear origin, and yet there are not energetic nuclear particles present in amounts commensurate with the energy produced [8]. If the large nuclear quantum can be fractionated, and the energy coherently coupled to a low energy mode (such as a phonon, plasmon, or perhaps magnon mode), then the physical mechanism would no longer be such a mystery.

The lossy spin–boson models that we have studied exhibit efficient coherent energy exchange under conditions where the large quantum is fractionated. We have proposed a generalization with two sets of two-level systems, in which one set is strongly coupled to the oscillator, and one set is weakly coupled to the oscillator (which we have termed a donor and receiver model) [9]. In such models, the initial excitation is in the weakly coupled two-level systems, which alone cannot fractionate the large two-level system quantum; however, when a strongly-coupled set of two-level systems is also present then the excitation from the weakly coupled systems is transferred to the strongly-coupled systems and fractionated.

We have for years considered this kind of model as a candidate to account for excess heat production in the Fleischmann–Pons experiment [10]. The mechanism that the model implements seems to be what is needed to account for the effect, and there seems to be indirect evidence in the two-laser experiment of Letts and Cravens [11] that the nuclear energy is communicated into specific optical phonon modes [12]. Another indirect connection to experiment is the observation that excess heat is correlated with a deuteron flux within the PdD, as one expects the deuteron flux to generate substantial incoherent optical phonon excitation (which is otherwise difficult to arrange for) [7]. In the donor and receiver models, no reactions occur unless the oscillator is highly excited, so we view the problem of triggering in these experiments in terms of developing strong excitation in high frequency vibrational modes [10].

It seems straightforward to identify the weakly-coupled donor transition in the model with $D_2/^4He$ transitions in the PdD, since the associated matrix element will be small because of tunneling through the Coulomb barrier. However, there have been difficulties for many years in the identification of the strongly-coupled receiver transition. While there has been no lack of candidate transitions, when we analyzed obvious candidates we found that the coupling was too weak to fractionate the large 24 MeV quantum from the donor transition under conditions relevant to experiment [10].

Earlier this year we recognized the existence of a relativistic coupling between lattice vibrations and internal nuclear degrees of freedom that results in a much stronger phonon exchange interaction [13]. This new interaction is important since it predicts a much stronger coupling than can be obtained from indirect electron-nuclear interactions; consequently, we have been optimistic that with this stronger coupling we might be able to finally identify receiver transitions involved in the excess heat experiments. The strongest coupling produced by the new interaction is found in the case of very highly excited nuclear excitation in which the upper state is extremely unstable. Since the donor and receiver model developed previously is based on strongly coupled receiver transitions with stable upper states, we cannot use it to model these transitions with the strongest coupling. To make progress, we need to revisit the basic lossy spin–boson model, and to repeat or extend the analysis for the special case where the upper states are unstable. This is the basic task we address in this work.

The lossy spin–boson model with unstable upper states is fundamentally different than the basic lossy spin–boson model that we analyzed before. Since the upper states are unstable, there cannot be net (real) excitation of the levels; hence, there is no evaluation of the coherent energy exchange rate in this case. Instead, the highly-exited oscillator causes a dynamical polarization of the two-level transitions, somewhat analogous to the dynamical polarization one would expect for a hydrogenic $1s - 2p$ transition in a slowly varying electric field (in this case, there is a mixing of the $1s$ and $2p$ states, but there is no real excitation of the $2p$ state). The analogy is not quite precise, since in our model there is an additional loss effect due to the coupling of the oscillator to fast decay channels available for states far off of resonance. In what follows we obtain results for two-level systems, for three-level systems, and for the generalization to N-level systems.

Armed with these results, we are in a position to return to the donor and receiver model which we can extend now to the case of receivers with unstable upper states. We find that the receiver is still able to fractionate a large quantum, at least in principle, and we obtain results for the donor dynamics in the presence of the coupled oscillator and receiver system. The subdivision effect that was present with stable receiver upper states is no longer present in the new model; without stable upper states the receiver must fractionate the large quantum completely. Without the ability to subdivide the large donor quantum, the requirements on the receiver transition are sufficiently severe that it is not possible to find receiver transitions (even with the new relativistic coupling) that can fractionate a 24 MeV quantum under conditions relevant to experiment.

Unfortunately, this result ends up ruling out the basic donor and receiver model as a candidate to account for excess heat in the Fleischmann–Pons experiment. This was disconcerting; we found this result while preparing for a conference (ICCF17), where we wanted to present more positive results. This provided the motivation for seeking some kind of modification of the model which might be more promising. The result of this work was the development of a new kind of model, one which appears to be in agreement with experiment in the cases we have studied so far, and which came into existence just in time to present at ICCF17 [14,15]. The lossy two-level model with unstable upper states considered in this work provides the foundation for this new model.

2. Model

We turn our attention now to a brief discussion of the new basic model. Our immediate goal then is to specify a Hamiltonian that implements a lossy spin–boson model with unstable upper states. However, it seems useful to begin the discussion at an earlier point in order to make the arguments more accessible.

2.1. Basic spin–boson model

We begin with the basic spin–boson model [16–18], which can be written as

$$\hat{H}_{\text{spin-boson}} \;=\; \Delta E \frac{\hat{S}_z}{\hbar} + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V(\hat{a}+\hat{a}^\dagger)\frac{2\hat{S}_x}{\hbar}. \tag{1}$$

This model includes a set of identical two-level systems with transition energy ΔE and an oscillator with characteristic energy $\hbar\omega_0$, linearly coupled with a coupling strength V. This model has been studied extensively in the literature, and we know that it describes coherent energy exchange between the two-level system and oscillator under conditions where the dressed two-level system energy is an odd multiple of the oscillator energy. The coherent energy exchange effect is weak in the multiphoton regime, and we have discussed previously that this is due to a destructive interference effect [1].

2.2. Lossy spin–boson model

Coherent energy exchange is very much faster in a generalization of the lossy spin–boson model when oscillator loss is present in the vicinity of the two-level system transition energy. We have written for this model the Hamiltonian

$$\hat{H}_{\text{lossy spin-boson}} \;=\; \Delta E \frac{\hat{S}_z}{\hbar} + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V(\hat{a}+\hat{a}^\dagger)\frac{2\hat{S}_x}{\hbar} - i\frac{\hbar\hat{\Gamma}(E)}{2}, \tag{2}$$

where the $i\hbar\hat{\Gamma}(E)/2$ term accounts for the loss in an infinite-order Brillouin–Wigner formalism (described in [1]). We call this model the lossy spin–boson model. There are issues connected to the inclusion of loss in this formalism that were raised by a reviewer; in response we have added an Appendix that discusses such issues.

We have been interested in the model in the strong coupling limit when a large number of two-level systems are present, since in this regime the coherent exchange rate between the two-level systems and oscillator can be reasonably large even when the two-level system quantum is fractionated into a great many oscillator quanta. The model looks deceptively simple, so that one might not anticipate the substantial amount of effort and approximations needed to determine the coherent energy exchange rate. Nevertheless, the coherent energy exchange rate has been determined as discussed in [2–4,19].

2.3. Lossy spin–boson model with an unstable upper state

In view of the discussion in the Introduction, we would like now to consider an extension of the lossy spin–boson model in which the upper state is unstable. We begin by first writing the lossy spin–boson model using a matrix notation

$$\hat{H}_{\text{lossy spin-boson}} = \frac{\Delta E}{2}\sum_j \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}_j + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V(\hat{a}+\hat{a}^\dagger)\sum_j \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}_j - i\frac{\hbar\hat{\Gamma}(E)}{2} \tag{3}$$

and then adding upper state loss to obtain

$$\hat{H} = \frac{\Delta E}{2}\sum_j \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}_j - i\frac{\hbar\hat{\gamma}(E)}{2}\sum_j \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}_j + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V(\hat{a}+\hat{a}^\dagger)\sum_j \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}_j - i\frac{\hbar\hat{\Gamma}(E)}{2}. \tag{4}$$

In this equation $\hat{\gamma}(E)$ models the decay of the unstable upper state of the two-level system, again within the infinite-order Brillouin–Wigner formalism. We presume in this that the two-level systems remain identical, and the decay rate is determined by the available system energy E consistent with decay processes in the Brillouin–Wigner approach.

2.4. Discussion

This provides us with a starting point for the analysis and extensions that will follow. This basic model by itself does not do very much; for example, we expect a mixing of the two-level and oscillator degrees of freedom that increases with coupling strength; but in general we are not looking for the mixing to lead to significant excitation of the upper state in order to see upper state decay. Our goal in this version of the model is to describe the basic mixing of the degrees of freedom, but otherwise the coupled system just sits there and doesn't do much that seems interesting.

What is interesting about the systems under discussion in this paper is that they provide a more relevant model for the physical systems that we are ultimately interested in. They behave somewhat differently than the lossy spin–boson models considered previously. Before moving on to use them in applications, we need to spend some time analyzing them in order to understand them.

3. Expansion Coefficients

To make progress generally on the problems of interest described in the Introduction we require approximate solutions for the eigenfunctions of the model, especially in the strong coupling regime. In spite of the deceptive simplicity of the basic model, there are subtle issues associated with the eigenfunctions, and exact analytic solutions are not available. We are going to have to make use of approximate and numerical solutions, and these will be pursued most easily if we work in terms of expansion coefficients $c_{m,n}$ in connection with the wavefunction construction

$$\Psi = \sum_m \sum_n c_{m,n} |S, m\rangle |n\rangle. \tag{5}$$

We recognize the $|S, m\rangle$ states as Dicke states associated with the pseudospin formalism, and $|n\rangle$ are the eigenstates of the simple harmonic oscillator.

3.1. Eigenvalue equation

We assume that Ψ satisfies the time-independent Schrödinger equation

$$E\Psi = \hat{H}\Psi. \tag{6}$$

The expansion coefficients then satisfy an eigenvalue equation, which we will approximate by

$$\begin{aligned} Ec_{m,n} = & \left(\Delta E m + \hbar\omega_0 n - i\frac{\hbar}{2}\Gamma(E)\right)c_{m,n} + V\sqrt{n+1}\sqrt{(S-m)(S+m-1)}c_{m+1,n+1} \\ & + V\sqrt{n}\sqrt{(S-m)(S+m-1)}c_{m+1,n-1} + V\sqrt{n+1}\sqrt{(S+m)(S-m+1)}c_{m-1,n+1} \\ & + V\sqrt{n}\sqrt{(S+m)(S-m+1)}c_{m-1,n-1}. \end{aligned} \tag{7}$$

What is missing in this explicit eigenvalue equation is the contribution of the upper state decay; we will take the point of view that the upper state decay is infinitely fast where it occurs, and exclude states from the problem that are unstable (consistent with the associated occupation probability vanishing). This greatly simplifies the problem.

3.2. Limit of large S and n

We are interested in the system when there are many two-level systems, and in general the polarization of the two-level systems will be weak. Under these conditions we may take

$$(S - m + 1) \rightarrow (S - m) \rightarrow 2S. \tag{8}$$

In addition, we assume that the oscillator is highly excited. The associated notation deserves some comment, as it is perhaps more iconic than mathematical. We assume that the oscillator is excited around some large number of quanta n_0, so that we might write

$$n = n_0 + \delta n. \tag{9}$$

For n sufficiently large we might write

$$\sqrt{n} \rightarrow \sqrt{n_0}, \qquad \sqrt{n+1} \rightarrow \sqrt{n_0}, \tag{10}$$

which removes the n-dependence from the coupling terms. The basis state energies depends on n, so we might write

$$\hbar\omega_0 n \rightarrow \hbar\omega_0(n_0 + \delta n). \tag{11}$$

All of this would suggest that we should write the eigenvalue equation as

$$\begin{aligned} Ec_{m,n_0+\delta n} \;=\; & \left(\Delta E m + \hbar\omega_0(n_0+\delta n) - i\frac{\hbar}{2}\Gamma(E)\right)c_{m,n_0+\delta n} \\ & + V\sqrt{n_0}\sqrt{2S}\Bigg[\sqrt{S+m-1}\left(c_{m+1,n_0+\delta n+1} + c_{m+1,n_0+\delta n-1}\right) \\ & + \sqrt{S+m}\left(c_{m-1,n_0+\delta n-1} + c_{m-1,n_0+\delta n-1}\right)\Bigg]. \end{aligned} \tag{12}$$

Such an eigenvalue equation could be understood from a mathematical point of view directly. However, we would like to work with a simpler notation. Since $\hbar\omega_0 n_0$ is a constant, we can eliminate it with no change in the dynamics (but we should remember that the energy eigenvalue would then be shifted). The $n_0 + \delta n$ appearing in the subscripts will be painful to deal with, so we replace them with n.

In the end, we work with an eigenvalue equation of the form

$$\begin{aligned} Ec_{m,n} \;=\; & \left(\Delta E m + \hbar\omega_0 n - i\frac{\hbar}{2}\Gamma(E)\right)c_{m,n} + V\sqrt{n_0}\sqrt{2S} \\ & \times\left[\sqrt{S+m-1}\left(c_{m+1,n+1} + c_{m+1,n-1}\right) + \sqrt{S+m}\left(c_{m-1,n-1} + c_{m-1,n-1}\right)\right]. \end{aligned} \tag{13}$$

In this equation we now think of n as incremental ($\delta n \to n$), and we have decided to go a bit against the literature and maintain the n_0 in the square root. Hopefully with this explanation and this notation things may be less confusing.

3.3. Dimensionless coupling constant

The dimensionless coupling constant that seems natural for this problem is

$$g \;=\; \frac{V\sqrt{n_0}\sqrt{2S}}{\Delta E}. \tag{14}$$

This dimensionless coupling constant differs from that of the spin–boson model written using a similar notation [20]

$$g_{\text{spin}-\text{boson}} \;=\; \frac{V\sqrt{n_0}}{\Delta E} \tag{15}$$

and also from what we have been using for the lossy spin–boson model [3]

$$g_{\text{lossy spin}-\text{boson}} \;=\; \frac{V\sqrt{n_0}\sqrt{S^2-m^2}}{\Delta E}. \tag{16}$$

It is important to note that big difference between the basic (lossless) spin–boson model and the lossy variants we have been interested in is that when loss is introduced the model responds to a dimensionless coupling constant that is greatly increased when many two-level systems are present.

The difference between the earlier lossy spin–boson model and the extension to the unstable upper state case looks to be drastic (going from $\sqrt{S^2-m^2}$ to $\sqrt{2S}$), but this would be misleading. There is a close connection between the earlier lossy spin–boson model and this new model, and the new one would behave very similarly to the old one in the vicinity of $m \approx -S$. So, writing the dimensionless coupling constant in this way here is more a matter of convenience, and it emphasizes that we are working with problems in which all of the two-level systems are close to being in ground states. When there is substantial excitation in the new model, the average coupling strength between individual states will be much greater than $V\sqrt{n_0}\sqrt{2S}/\Delta E$, and will be much closer instead to $V\sqrt{n_0}\langle\sqrt{S^2-m^2}\rangle/\Delta E$. Unfortunately it will be inconvenient to work with a dimensionless coupling constant that is implicitly defined in terms of itself. For a given solution, we will be able to determine what the suitable averaged dimensionless coupling strength is if we would like to compare with the earlier lossy spin–boson model.

With this definition we may write the normalized eigenvalue equation as

$$\epsilon c_{m,n} = \left(m + \frac{n}{\Delta n} - i\frac{\hbar}{2\Delta E}\Gamma(\epsilon)\right)c_{m,n} + g\left[\sqrt{S+m-1}\left(c_{m+1,n+1} + c_{m+1,n-1}\right) + \sqrt{S+m}\left(c_{m-1,n-1} + c_{m-1,n-1}\right)\right], \tag{17}$$

where

$$\epsilon = \frac{E}{\Delta E}. \tag{18}$$

3.4. Loss and boundary conditions

In some of our early studies with the lossy spin–boson model we included explicit loss models that included estimates of the decay rates as a function of energy. We found in general that the probability amplitude tended to avoid states with high loss, and that when the dimensionless coupling constant became large, that the boundary appeared sharp on the scale of the overall probability amplitude as a function of m and n. Consequently, when we carried out detailed analyses relevant to the strong coupling limit, it seemed natural to work with an "infinite loss" version of the model in which we simply omitted unstable states [2]. Consistent with the premise of such a model, all basis states with energies below some threshold would be removed from the calculation as a way to approximately model the effect of loss.

We encounter this basic issue anew in the new version of the model with unstable upper states. Since this same loss is present in the new model, we would make a similar approximation by excluding all basis states below a fixed energy. But now we have an additional loss effect in which we need to exclude states that involve real occupation of excited states; this is new and deserves some thought.

Coherent energy exchange in the previous model resulted in real excited states, in which Δn oscillator quanta were lost and one unit of excitation $\Delta E = \hbar\omega\Delta n$ was gained. In the new model this cannot occur since all real excited states are unstable. But suppose that we were to consider states in which less than Δn oscillator quanta were lost, so that an excited state could be formed off of resonance (as a virtual state). Depending on the loss channels available, we would probably still expect it to decay rapidly, although not as rapidly as if it had its full energy. Based on this picture, we should exclude such states as well.

The end result of such a line of argument is that we might implement upper state decay approximately by omitting basis states with an oscillator energy below some threshold value. If so, we would write the normalized eigenvalue equation as

$$\epsilon c_{m,n} = \left(m + \frac{n}{\Delta n}\right)c_{m,n} + g\left[\sqrt{S+m-1}\left(c_{m+1,n+1} + c_{m+1,n-1}\right) + \sqrt{S+m}\left(c_{m-1,n-1} + c_{m-1,n-1}\right)\right] \tag{19}$$

subject to the boundary condition

$$c_{m,n} \;=\; 0, \qquad n < 0 \tag{20}$$

taking the incremental n to be zero at the cut off.

3.5. Solutions in the weak coupling regime

Based on the discussion above we have arrived at a model that can be analyzed, and one of our first tasks is to consider which of the many possible eigenfunction solutions are of interest. Consider first the situation where the dimensionless coupling strength is small; for example, suppose that

$$\Delta n \;=\; \frac{\Delta E}{\hbar\omega_0} \;=\; 1000, \tag{21}$$

$$g \;=\; 0.03. \tag{22}$$

We can compute solutions numerically, and categorize them usefully by plotting out values of $\langle m+S\rangle$ and $\langle n\rangle$ as shown in Fig. 1. Since the coupling is weak for this example, for the most part we do not expect much to happen. We see that most of the states have $\langle m+S\rangle$ and $\langle n\rangle$ averages which closely match the basis state equivalents. The biggest impact from the coupling can be seen in the lower left corner, where there is some mixing between basis states with $m+S=0$ and with $n=0$ and $n=2$.

All of the states indicated to the right of the first column involve $\langle m+S\rangle$ values on the order of 1 or higher; these states clearly involve one or more excited two-level systems. As such, we would not make use of them for our model, since excited states are unstable.

Based on this, all of the solutions indicated in the first column then are candidate wavefunctions that we might consider using to describe our coupled system. But how do we think about them? We know that the state at the bottom is closest to the cut off, so it will be most impacted by the effect of the boundary. We would expect that once we have gone up the column far enough that we will find states sufficiently removed from the boundary that the boundary will have no impact. The choice of one such state over another in the weak coupling regime appears to then be connected with where the cut off in oscillator quanta is relative to the basis states that make up the eigenfunction. If the coupling is weak this can make a substantial difference, as we can see already in the shifted $\langle n\rangle$ value of the state closest to the boundary.

3.6. Solutions when the coupling is stronger

The situation changes qualitatively when the coupling is stronger. Perhaps the best way to see this simply is to consider the lowest two eigenfunctions when the coupling is not so weak. Consider an example in which

$$\Delta n \;=\; \frac{\Delta E}{\hbar\omega_0} \;=\; 100, \tag{23}$$

$$g \;=\; 0.70. \tag{24}$$

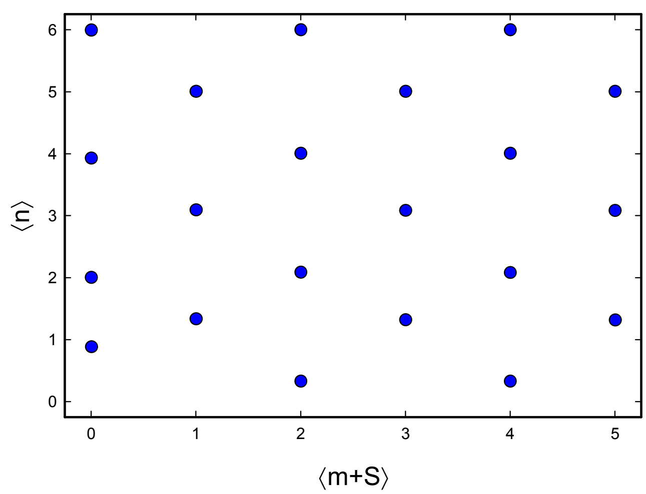

Figure 1. Average values $\langle m + S \rangle$ and $\langle n \rangle$ for low-lying states with even $n + m + S$ basis states in weak coupling.

The lowest two eigenfunctions are shown in Figs. 2 and 3. We see that the situation seems qualitatively different now; these eigenfunctions closest to the boundary are showing a collective effect beyond primarily translation and simple mixing as in the weak coupling limit. We see in these eigenfunctions a ground state and single unit of collective excitation in n, and there is also a solution which shows a similar single unit of excitation in $m + S$. We will be interested in these solutions in a following paper where they will constitute a set of off-resonant intermediate states that will contribute in a coherent dynamics calculation.

3.7. Intermediate regime

We recognize in this discussion that there should occur an intermediate regime. One intermediate regime we might associate with values of g in which the system changes over from the weak coupling regime to the strong coupling regime; another intermediate regime would be expected when the spread in oscillator quanta leads to occupation of states that decay with associated rates that are slow or moderate. The development of models relevant to these regimes will be interesting, especially in the latter case; we consider such endeavors to be outside of the scope of this study.

3.8. Hard and soft boundary conditions

In our earlier work on the lossy spin–boson model we noticed an issue with respect to the "hard" boundary condition discussed above. As a mathematical statement it seems clear that we might draw a line at some value of n and exclude basis states below the line. This is similar to the assumption that we made in our earlier work where we excluded basis states below a fixed energy. We found in that case that the wavefunctions that result were finite at the threshold energy [4].

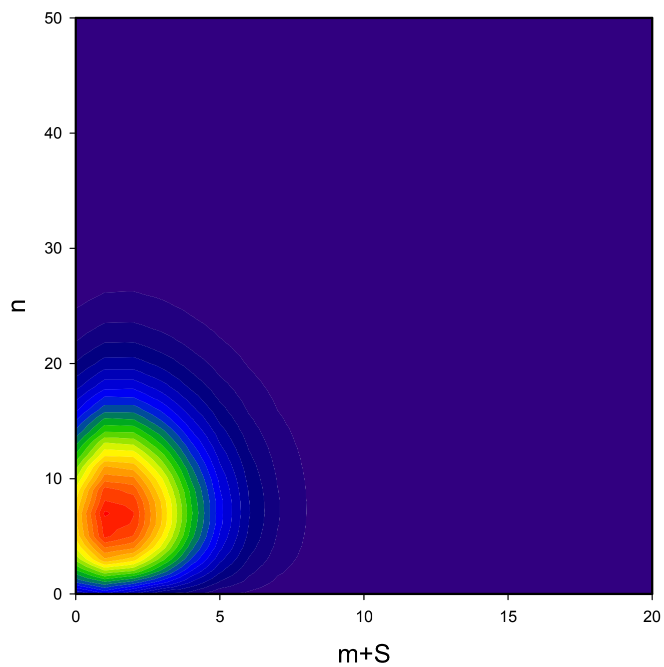

Figure 2. Contour plot of the lowest eigenfunction solution plotted as $(-1)^m c_{m,n}$ as a function of $m+S$ and n.

Since there has been some time since this earlier work, we have had time to think about this assumption and the consequences. One unexpected consequence is that it leads to a discontinuous slope for zero energy exchange in the line shape we compute for the broad x-ray feature in the Karabut experiment. This motivated us to reconsider the boundary condition, and prompts us here to propose a modified boundary condition which we may write as

$$c_{m,0} = 0 \tag{25}$$

with excluded states for $n < 0$. The idea is that when decay processes occur as the excitation of the lattice decreases, the effect will be gradual with n; we would not expect in the physical system that there will be no decay above some threshold value of n and infinite decay below that threshold.

4. Approximation with Product Wavefunction

It is of interest to see whether an approximate product wavefunction might be useful for this problem, as it would allow us to analyze the strong coupling regime more easily. To proceed, we approximate the expansion coefficients using

$$c_{m,n} = (-1)^m a_m u_n. \tag{26}$$

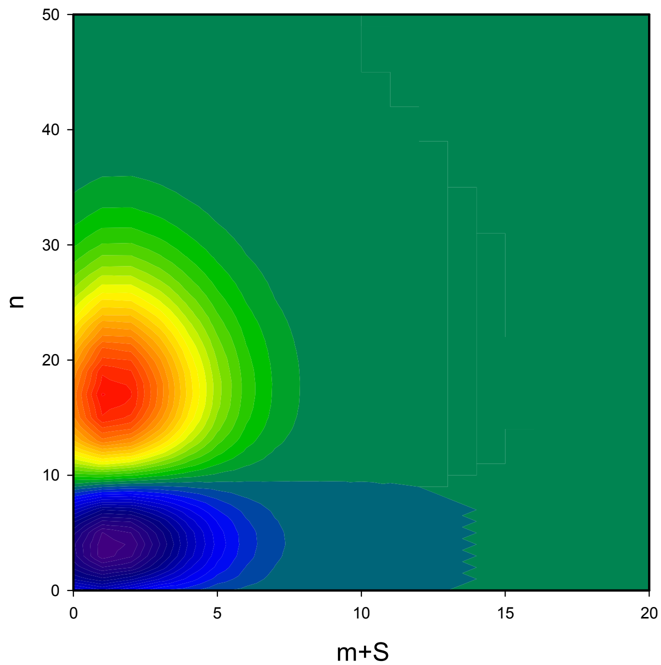

Figure 3. Contour plot of the first excited state plotted as $(-1)^m c_{m,n}$ as a function of $m + S$ and n.

This is analogous the the pulse and amplitude approximation that we pursued in the case of the lossy spin–boson model [21].

4.1. Variational model

The eigenvalue equation for the expansion coefficients can be derived from a variational principle based on

$$I = \sum_{m,n}\left(m+\frac{n}{\Delta n}\right)c_{m,n}^2 - g\sum_{m,n}c_{m,n}\left[\sqrt{S+m-1}\left(c_{m+1,n+1}+c_{m+1,n-1}\right) + \sqrt{S+m}\left(c_{m-1,n-1}+c_{m-1,n-1}\right)\right] \tag{27}$$

subject to the constraint

$$\sum_{m,n}c_{m,n}^2 = 1. \tag{28}$$

This motivates us to propose a modified variational principle for the approximate product wavefunction

$$J = \sum_m m a_m^2 + \frac{1}{\Delta n}\sum_n n u_n^2 - g\left[\sum_n u_n(u_{n+1}+u_{n-1})\right] \times \left[\sum_m\left(\sqrt{S+m-1}\,a_m a_{m+1} + \sqrt{S+m}\,a_m a_{m-1}\right)\right] \tag{29}$$

subject to the constraints

$$\sum_m a_m^2 = 1, \tag{30}$$

$$\sum_n u_n^2 = 1. \tag{31}$$

4.2. Optimization of the product wavefunction

We can make J stationary if a_m satisfies the constraint

$$\lambda_a a_m = m a_m - g\left[\sum_n u_n(u_{n+1}+u_{n-1})\right]\left(\sqrt{S+m-1}\,a_{m+1} + \sqrt{S+m}\,a_{m-1}\right) \tag{32}$$

with the boundary conditions

$$a_m = 0 \qquad \text{for } m < -S, \tag{33}$$

$$a_m \to 0 \qquad \text{for } m \to \infty. \tag{34}$$

In addition, the other function u_n satisfies the constraint

$$\lambda_u u_n = \frac{n}{\Delta n}u_n - g\left[\sum_m\left(\sqrt{S+m-1}\,a_m a_{m+1} + \sqrt{S+m}\,a_m a_{m-1}\right)\right](u_{n+1}+u_{n-1}) \tag{35}$$

subject to

$$u_n = 0 \qquad \text{for } n < 0, \tag{36}$$

$$u_0 = 0, \tag{37}$$

$$u_n \to 0 \qquad \text{for } n \to \infty. \tag{38}$$

4.3. Comparison

We have carried out computations with the product solution in order to compare with the exact solution for the parameters

$$\Delta n = \frac{\Delta E}{\hbar\omega_0} = 70, \tag{39}$$

$$g = 1.80. \tag{40}$$

We see from Figs. 4 and 5 that the optimized product solution is quite close to the exact numerical solution.

5. Sets of N-level Systems Coupled to an Oscillator

We are also interested in more complicated models than those discussed in the earlier sections. The simplest example of a nucleus interacting with a lattice in the strong coupling regime is the case of internal excitation of the deuteron coupled to highly excited optical phonon modes in PdD. In this case there are three spin states associated with the deuteron, two of which can undergo transitions to different upper states [22]. Most other examples involve coupling to many excited states (transitions in Pd or Ni), and also involve transitions in different isotopes. This provides us with motivation to examine more complicated versions of the model.

To reduce the complexity of the problem that results, we will restrict our attention to two specific issues in what follows: multiple excited states and different interacting systems. In doing so we defer the problem of multiple ground states interacting with common excited states, which makes things substantially more complicated. Our approach will be to focus first on the interaction of two three-level systems (with a single ground state and two unstable excited states) with a highly excited oscillator, as before in the limit that the transition energies are much greater than the characteristic oscillator energy. The analysis of this problem will allow us to generalize in a straightforward way to the case of many N-level systems interacting with a common oscillator, under the restriction that each has only a single ground state.

5.1. Model Hamiltonian

We can write a Hamiltonian for two sets of three-level systems interacting with an oscillator as

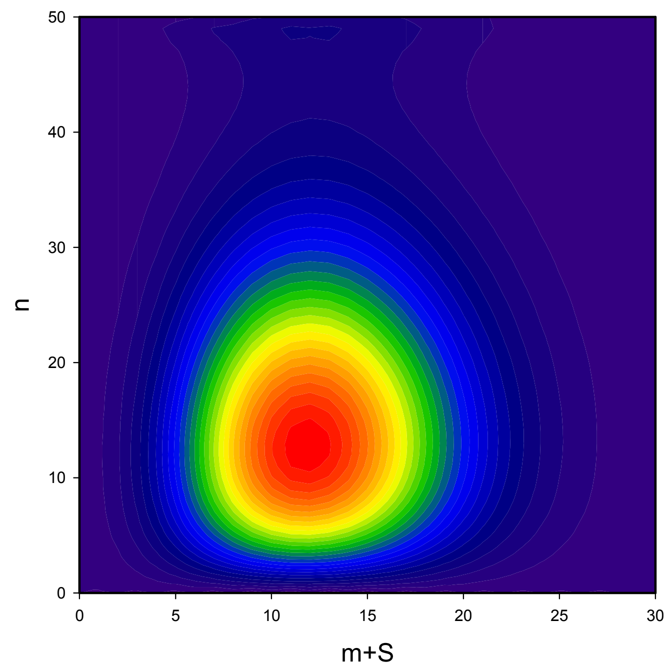

Figure 4. Exact lowest eigenfunction solution plotted as $c_{m,n}$ as a function of $m+S$ and n.

$$
\begin{aligned}
\hat{H} \;=\; & \sum_j \begin{pmatrix} E_1^{(1)} & 0 & 0 \\ 0 & E_2^{(1)} & 0 \\ 0 & 0 & E_3^{(1)} \end{pmatrix}_j + \sum_k \begin{pmatrix} E_1^{(2)} & 0 & 0 \\ 0 & E_2^{(2)} & 0 \\ 0 & 0 & E_3^{(2)} \end{pmatrix}_k + \hbar\omega_0 \hat{a}^\dagger \hat{a} \\
& + \sum_j \begin{pmatrix} 0 & V_{12}^{(1)} & V_{13}^{(1)} \\ V_{21}^{(1)} & 0 & V_{23}^{(1)} \\ V_{31}^{(1)} & V_{32}^{(1)} & 0 \end{pmatrix}_j (\hat{a}+\hat{a}^\dagger) + \sum_k \begin{pmatrix} 0 & V_{12}^{(2)} & V_{13}^{(2)} \\ V_{21}^{(2)} & 0 & V_{23}^{(2)} \\ V_{31}^{(2)} & V_{32}^{(2)} & 0 \end{pmatrix}_k (\hat{a}+\hat{a}^\dagger) \\
& - \frac{i\hbar}{2} \sum_j \begin{pmatrix} 0 & 0 & 0 \\ 0 & \hat{\gamma}_2^{(1)}(E) & 0 \\ 0 & 0 & \hat{\gamma}_3^{(1)}(E) \end{pmatrix}_j - \frac{i\hbar}{2} \sum_k \begin{pmatrix} 0 & 0 & 0 \\ 0 & \hat{\gamma}_2^{(2)}(E) & 0 \\ 0 & 0 & \hat{\gamma}_3^{(2)}(E) \end{pmatrix}_k - i\frac{\hbar\hat{\Gamma}(E)}{2}.
\end{aligned}
\tag{41}
$$

Note that the notation appropriate for the three-level systems in this case is reversed relative to that of the two-level systems. One set of three-level systems is located at a set of sites denoted by the index j, and the other set of three-level systems is located at sites denoted by k. Single-phonon exchange transitions are included for both sets of three-level

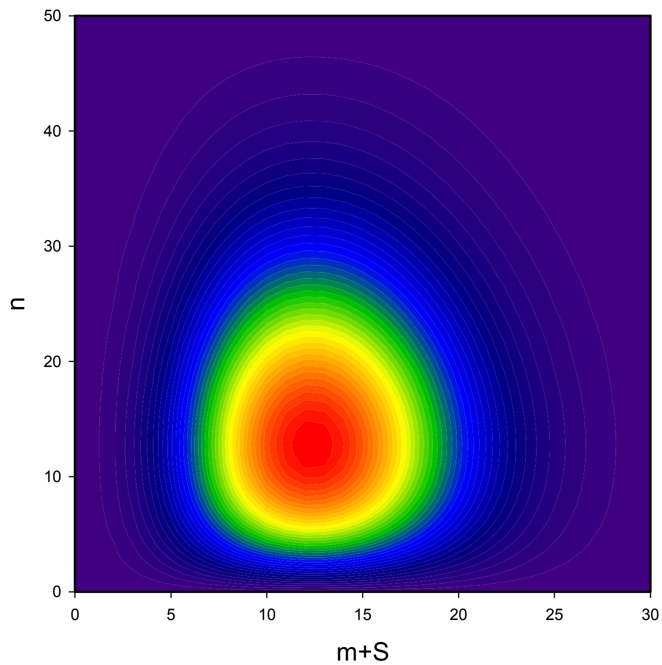

Figure 5. Approximate product lowest eigenfunction solution plotted as $c_{m,n}$ as a function of $m + S$ and n.

systems. Oscillator loss is included as before through a Brillouin–Wigner loss operator $-i\hbar\hat{\Gamma}(E)/2$; and we indicate the loss associated with the unstable upper states through appropriate $\hat{\gamma}(E)$ operators.

5.2. Expansion coefficients

This model is more complicated than the extension of the lossy spin–boson model considered in previous sections; however, we can reduce the complexity some by working with expansion coefficients. We assume a wavefunction for the coupled system of the form

$$\Psi = \sum_n \sum_{N_2^{(1)}} \sum_{N_3^{(1)}} \sum_{N_2^{(2)}} \sum_{N_3^{(2)}} c_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} \left|N_1^{(1)}, N_2^{(1)}, N_3^{(1)}\right\rangle \left|N_1^{(2)}, N_2^{(2)}, N_3^{(2)}\right\rangle |n\rangle, \tag{42}$$

where the different $|N_1, N_2, N_3\rangle$ states are the generalization of Dicke states for the two three-level systems.

We are interested in developing solutions to the time-independent Schrödinger equation

$$E\Psi = \hat{H}\Psi. \tag{43}$$

In the case of the basis state expansion above this leads to an eigenvalue equation for the expansion coefficients which is of the form

$$\begin{aligned}
E\, c_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} = & \Big[E_1^{(1)}N_1^{(1)} + E_2^{(1)}N_2^{(1)} + E_3^{(1)}N_3^{(1)} \\
& + E_1^{(2)}N_1^{(2)} + E_2^{(2)}N_2^{(2)} + E_3^{(2)}N_3^{(2)} + n\hbar\omega_0\Big] c_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} \\
& + V_{12}^{(1)}\sqrt{N_1^{(1)}(N_2^{(2)}+1)}\Big[\sqrt{n+1}\,c_{N_1^{(1)}+1,N_2^{(1)}-1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n+1} \\
& + \sqrt{n}\,c_{N_1^{(1)}+1,N_2^{(1)}-1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n-1}\Big] \\
& + V_{21}^{(1)}\sqrt{(N_1^{(1)}+1)N_2^{(2)}}\Big[\sqrt{n+1}\,c_{N_1^{(1)}-1,N_2^{(1)}+1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n+1} \\
& + \sqrt{n}\,c_{N_1^{(1)}-1,N_2^{(1)}+1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n-1}\Big] \\
& + \cdots,
\end{aligned} \tag{44}$$

where the $\cdots$ denotes a very large number of interaction terms similar to the ones included, and where we include loss through appropriate boundary conditions as above.

5.3. Limit of large n, large $N_1^{(1)}$ and $N_1^{(2)}$

Even though we are interested in the strong coupling regime, we expect the occupation of the ground state to dominate for both sets of three-level systems. Since the interaction terms are present proportional to the appropriate Dicke factors (such as $\sqrt{(N_1^{(1)}+1)N_2^{(2)}}$) we see that interactions with the ground states will be favored. This suggests that we might simplify things by retaining only interactions involving ground states, and using the large N approximations

$$\sqrt{N_1+1} \rightarrow \sqrt{N_1}, \tag{45}$$

$$\sqrt{n+1} \rightarrow \sqrt{n_0}. \tag{46}$$

In addition, it will be convenient to adopt a phase convention in which all of the coupling matrix elements from ground states are real, so

$$V_{12} = V_{21} \quad \text{(real)}, \tag{47}$$

$$V_{13} = V_{31} \quad \text{(real)}. \tag{48}$$

This leads to an approximate eigenvalue equation of the form

$$
\begin{aligned}
E\, c_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} = & \Big[E_2^{(1)} N_2^{(1)} + E_3^{(1)} N_3^{(1)} \\
& + E_2^{(2)} N_2^{(2)} + E_3^{(2)} N_3^{(2)} + n\hbar\omega_0\Big] c_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} \\
& + V_{12}^{(1)} \sqrt{n_0} \sqrt{N_1^{(1)}} \Big\{ \sqrt{N_2^{(2)}+1} \Big[c_{N_1^{(1)}+1,N_2^{(1)}-1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n+1} \\
& + c_{N_1^{(1)}+1,N_2^{(1)}-1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n-1} \Big] \\
& + \sqrt{N_2^{(2)}} \Big[c_{N_1^{(1)}-1,N_2^{(1)}+1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n+1} \\
& + c_{N_1^{(1)}-1,N_2^{(1)}+1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n-1} \Big] \Big\} \\
& + \cdots ,
\end{aligned} \tag{49}
$$

where we have taken the ground state energy E_1 for the two three-level systems to be zero.

5.4. Approximate product solution

Based on the utility of the approximate product solution described above, we would like to generalize it to the present case. We consider an approximate product solution of the form

$$
c_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} = (-1)^{N_1^{(1)}+N_2^{(1)}+N_1^{(2)}+N_2^{(2)}} a^{(1)}_{N_2^{(1)}} b^{(1)}_{N_3^{(1)}} a^{(2)}_{N_2^{(2)}} b^{(2)}_{N_3^{(2)}} u_n . \tag{50}
$$

There are selection rules for the expansion coefficients (half of them are zero) as was the case for the two-level version of the problem considered above. Within the product wavefunction approximation we lose the selection rule, but we face no difficulties with this in applications later on. It is possible to impose selections rule once the various terms in the product wavefunction are optimized if needed.

5.5. Variational principle

To proceed, we would like to optimize the product solution based on a variational principle. We begin by considering the variational principle associated with the original expansion coefficients $c_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n}$. We write

$$
\begin{aligned}
I \;=\; & \sum_n \sum_{N_2^{(1)}} \sum_{N_3^{(1)}} \sum_{N_2^{(2)}} \sum_{N_3^{(2)}} \Big[E_2^{(1)} N_2^{(1)} + E_3^{(1)} N_3^{(1)} \\
& + E_2^{(2)} N_2^{(2)} + E_3^{(2)} N_3^{(2)} + n\hbar\omega_0 \Big] c^2_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} \\
& + V_{12}^{(1)} \sqrt{n_0} \sqrt{N_1^{(1)}} \sum_n \sum_{N_2^{(1)}} \sum_{N_3^{(1)}} \sum_{N_2^{(2)}} \sum_{N_3^{(2)}} \Big\{ \sqrt{N_2^{(2)}+1}\, c_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} \\
& \Big[c_{N_1^{(1)}+1,N_2^{(1)}-1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n+1} + c_{N_1^{(1)}+1,N_2^{(1)}-1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n-1} \Big] \\
& + \sqrt{N_2^{(2)}}\, c_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} \\
& \Big[c_{N_1^{(1)}-1,N_2^{(1)}+1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n+1} + c_{N_1^{(1)}-1,N_2^{(1)}+1,N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n-1} \Big] \Big\} \\
& + \cdots
\end{aligned}
\tag{51}
$$

subject to

$$
\sum_n \sum_{N_2^{(1)}} \sum_{N_3^{(1)}} \sum_{N_2^{(2)}} \sum_{N_3^{(2)}} c^2_{N_1^{(1)},N_2^{(1)},N_3^{(1)},N_1^{(2)},N_2^{(2)},N_3^{(2)},n} \;=\; 1
\tag{52}
$$

Based on this, we consider the optimization of the product wavefunction based on

$$
\begin{aligned}
J \;=\; & \sum_{N_2^{(1)}} E_2^{(1)} [a^{(1)}_{N_2^{(1)}}]^2 + \sum_{N_3^{(1)}} E_3^{(1)} [b^{(1)}_{N_3^{(1)}}]^2 + \sum_{N_2^{(2)}} E_2^{(2)} [a^{(2)}_{N_2^{(2)}}]^2 + \sum_{N_3^{(2)}} E_3^{(2)} [b^{(2)}_{N_3^{(2)}}]^2 + \sum_n \hbar\omega_0 n u_n^2 \\
& - V_{12}^{(1)} \sqrt{n_0} \sqrt{N_1^{(1)}} \sum_{N_2^{(1)}} a^{(1)}_{N_2^{(1)}} \Big(\sqrt{N_2^{(1)}+1}\, a^{(1)}_{N_2^{(1)}+1} + \sqrt{N_2^{(1)}}\, a^{(1)}_{N_2^{(1)}-1} \Big) \sum_n u_n \Big(u_{n+1} + u_{n-1} \Big) \\
& - V_{13}^{(1)} \sqrt{n_0} \sqrt{N_1^{(1)}} \sum_{N_3^{(1)}} b^{(1)}_{N_3^{(1)}} \Big(\sqrt{N_3^{(1)}+1}\, b^{(1)}_{N_3^{(1)}+1} + \sqrt{N_3^{(1)}}\, b^{(1)}_{N_3^{(1)}-1} \Big) \sum_n u_n \Big(u_{n+1} + u_{n-1} \Big) \\
& - V_{12}^{(2)} \sqrt{n_0} \sqrt{N_1^{(2)}} \sum_{N_2^{(2)}} a^{(2)}_{N_2^{(2)}} \Big(\sqrt{N_2^{(2)}+1}\, a^{(2)}_{N_2^{(2)}+1} + \sqrt{N_2^{(2)}}\, a^{(2)}_{N_2^{(2)}-1} \Big) \sum_n u_n \Big(u_{n+1} + u_{n-1} \Big) \\
& - V_{13}^{(2)} \sqrt{n_0} \sqrt{N_1^{(2)}} \sum_{N_3^{(2)}} b^{(2)}_{N_3^{(2)}} \Big(\sqrt{N_3^{(2)}+1}\, b^{(2)}_{N_3^{(2)}+1} + \sqrt{N_3^{(2)}}\, b^{(2)}_{N_3^{(2)}-1} \Big) \sum_n u_n \Big(u_{n+1} + u_{n-1} \Big)
\end{aligned}
\tag{53}
$$

subject to

$$
\sum_{N_2^{(1)}} [a^{(1)}_{N_2^{(1)}}]^2 \;=\; 1, \qquad \sum_{N_3^{(1)}} [b^{(1)}_{N_3^{(1)}}]^2 \;=\; 1,
\tag{54}
$$

$$\sum_{N_2^{(2)}}[a^{(2)}_{N_2^{(2)}}]^2 \ = \ 1, \qquad \sum_{N_3^{(2)}}[b^{(2)}_{N_3^{(2)}}]^2 \ = \ 1, \tag{55}$$

$$\sum_n u_n^2 \ = \ 1. \tag{56}$$

5.6. Optimization of the product wavefunction

We use the variational principle to optimize the approximate product wavefunction, and we obtain constraints of the form

$$\lambda_a a_{N_2} \ = \ E_2 a_{N_2} - V_{12}\sqrt{n_0}\sqrt{N_1}\Big[\sum_n u_n(u_{n+1}+u_{n-1})\Big]\Big[\sqrt{N_2+1}a_{N_2+1}+\sqrt{N_2}a_{N_2-1}\Big], \tag{57}$$

$$\lambda_b b_{N_3} \ = \ E_3 b_{N_3} - V_{13}\sqrt{n_0}\sqrt{N_1}\Big[\sum_n u_n(u_{n+1}+u_{n-1})\Big]\Big[\sqrt{N_3+1}b_{N_3+1}+\sqrt{N_3}b_{N_3-1}\Big], \tag{58}$$

where there are one of each constraint for the two different sets of three-level systems. In addition, we end up with a constraint for u_n of the form

$$\begin{aligned}\lambda_u u_n \ = \ \hbar\omega_0 u_n - \Bigg[& V_{12}^{(1)}\sqrt{n_0}\sqrt{N_1^{(1)}}\sum_{N_2^{(1)}} a^{(1)}_{N_2^{(1)}}\left(\sqrt{N_2^{(1)}+1}a^{(1)}_{N_2^{(1)}+1}+\sqrt{N_2^{(1)}}a^{(1)}_{N_2^{(1)}-1}\right) \\ & + V_{13}^{(1)}\sqrt{n_0}\sqrt{N_1^{(1)}}\sum_{N_3^{(1)}} b^{(1)}_{N_3^{(1)}}\left(\sqrt{N_3^{(1)}+1}b^{(1)}_{N_3^{(1)}+1}+\sqrt{N_3^{(1)}}b^{(1)}_{N_3^{(1)}-1}\right) \\ & + V_{12}^{(2)}\sqrt{n_0}\sqrt{N_1^{(2)}}\sum_{N_2^{(2)}} a^{(2)}_{N_2^{(2)}}\left(\sqrt{N_2^{(2)}+1}a^{(2)}_{N_2^{(2)}+1}+\sqrt{N_2^{(2)}}a^{(2)}_{N_2^{(2)}-1}\right) \\ & + V_{13}^{(2)}\sqrt{n_0}\sqrt{N_1^{(2)}}\sum_{N_3^{(2)}} b^{(2)}_{N_3^{(2)}}\left(\sqrt{N_3^{(2)}+1}b^{(2)}_{N_3^{(2)}+1}+\sqrt{N_3^{(2)}}b^{(2)}_{N_3^{(2)}-1}\right)\Bigg](u_{n+1}+u_{n-1}).\end{aligned} \tag{59}$$

We can implement the loss model in this case by omitting u_n for $n < 0$, and adopting the boundary condition

$$u_0 \ = \ 0. \tag{60}$$

5.7. Discussion

Although we started out with a rather complicated model, within the framework of the product wavefunction approximation we have obtained a set of constraints that are closely connected with what we obtained above for the two-level system version of the problem. In connection with each excited state we end up with a constraint of the form

$$\lambda_a a_N \; = \; \Delta E a_N - V\sqrt{n_0}\sqrt{N_1}\left[\sum_n u_n(u_{n+1}+u_{n-1})\right]\left[\sqrt{N+1}a_{N+1}+\sqrt{N}a_{N-1}\right]. \tag{61}$$

We expect this to be true even for systems more complicated than three-level systems, and also if there are more than two different sets.

In connection with the oscillator, we obtain a constraint that we might write as

$$\begin{aligned}\lambda_u u_n \; = \; & \hbar\omega_0 u_n - \left[\sum_\beta \sqrt{N_1^{(\beta)} n_0}\, \sum_\kappa V_{1\kappa}^{(\beta)} \right. \\ & \left. \sum_{N_\kappa^{(\beta)}} a^{(\beta)}_{N_\kappa^{(\beta)}}\left(\sqrt{N_\kappa^{(\beta)}+1}\; a^{(\beta)}_{N_\kappa^{(\beta)}+1} + \sqrt{N_\kappa^{(\beta)}}\; a^{(\beta)}_{N_\kappa^{(\beta)}-1}\right)\right](u_{n+1}+u_{n-1}).\end{aligned} \tag{62}$$

The spreading of the oscillator distribution in this model is determined by contributions from all of the different isotopes (denoted by β), with individual contributions over the different excited states of each isotope (denoted by κ). To obtain the contributions in each case, we need to sum over the appropriate excited state distribution (denoted by $N_\kappa^{(\beta)}$).

6. Approximation for the Oscillator Distribution

In the applications of the model that will follow, we will be most interested in estimates for the oscillator distribution given a particular set of nuclei and a specific excitation of the oscillator (in terms of n_0). From the discussion of the previous section we can solve self-consistently for the excited state distributions and oscillator distribution, although it requires some effort to do so.

It would be useful to develop simpler analytic estimates in connection with the product solution above in order to gain a better understanding of how the contributions of the different transitions in the various isotopes spread the oscillator distribution. We recognize two features of the constraints associated with the product model in the strong coupling limit that can lead to a simplification. In the strong coupling regime the oscillator distribution is slowly varying, and we can use this to isolate the different constraints that pertain to the excited state distribution. Additionally, we recognize that we do not actually need explicit solutions for the excited state distributions; instead we only need an estimate for the average off-diagonal transition matrix element. We can use these to develop a specific solution for the oscillator distribution directly from a knowledge of the transition parameters. We will pursue these ideas in what follows.

6.1. Isolation of the excited state distributions

In the strong coupling regime the oscillator distribution will be spread out over a great many oscillator states, and from earlier work we know that when a product wavefunction is used the u_n distribution will be slowly varying so that

$$u_{n\pm1} \; \approx \; u_n. \tag{63}$$

In this case we can write

$$\sum_n u_n(u_{n+1}+u_{n-1}) \; = \; 2\sum_n u_n^2 \; = \; 2. \tag{64}$$

Each constraint associated with the excited state distributions in this case can then be dealt with separately, and is described through

$$\lambda_a a_N \;=\; \Delta E a_N - 2V\sqrt{n_0}\sqrt{N_1}\Big[\sqrt{N+1}\,a_{N+1} + \sqrt{N}\,a_{N-1}\Big]. \tag{65}$$

This is potentially interesting, since the distribution ultimately depends on a single parameter; we can recast this constraint in the form

$$\lambda'_a a_N \;=\; a_N - 2g_a\Big[\sqrt{N+1}\,a_{N+1} + \sqrt{N}\,a_{N-1}\Big] \tag{66}$$

with $\lambda'_a = \lambda_a/\Delta E$, and where g_a is defined as

$$g_a \;=\; \frac{V\sqrt{n_0}\sqrt{N_1}}{\Delta E}. \tag{67}$$

6.2. Estimate for the contribution of each excited state

As a practical matter, we don't require the excited state distributions in order to solve for the oscillator distribution; instead, we require the sum

$$f(g_a) \;=\; \sum_N a_N\Big(\sqrt{N+1}\,a_{N+1} + \sqrt{N}\,a_{N-1}\Big). \tag{68}$$

This can be evaluated directly from a numerical solution of the constraint equation; we find

$$f(g_a) \;=\; 4g_a. \tag{69}$$

We can use this to write for the oscillator distribution

$$\lambda_u u_n \;=\; \hbar\omega_0 u_n - \Bigg[\sum_\beta\sum_\kappa \frac{4[V_{1\kappa}^{(\beta)}]^2 n_0 N_1^{(\beta)}}{E_\kappa^{(\beta)}}\Bigg](u_{n+1}+u_{n-1}). \tag{70}$$

We see that the oscillator distribution in this model depends only on a single parameter; we may write

$$\lambda'_u u_n \;=\; u_n - 2g_u(u_{n+1}+u_{n-1}), \tag{71}$$

where $\lambda'_u = \lambda_u/\hbar\omega_0$, and where the dimensionless coupling strength appropriate to the oscillator distribution is

$$g_u \;=\; \sum_\beta\sum_\kappa \frac{2[V_{1\kappa}^{(\beta)}]^2 n_0 N_1^{(\beta)}}{\hbar\omega_0 E_\kappa^{(\beta)}}. \tag{72}$$

In this summation β indicates the isotope and κ indicates the transition within the isotope.

6.3. Continuum approximation

This model will be most interesting to us in connection with coherent energy exchange under conditions that a great many oscillator quanta are exchanged. In this case the spread in the oscillator distribution will be very large, and this motivates us to pursue a continuum approximation. We can adopt a continuum approximation using

$$\text{discrete } n \;\rightarrow\; \text{continuous } n, \tag{73}$$

$$u_n \;\rightarrow\; u(n). \tag{74}$$

We can use this to write

$$u_{n+1} + u_{n-1} \;\rightarrow\; u(n+1) + u(n-1) \;=\; 2u(n) + \frac{\mathrm{d}^2}{\mathrm{d}n^2}u(n) + \cdots \tag{75}$$

The continuous version of the eigenvalue equation becomes

$$\lambda' u(n) \;=\; nu(n) - 4g_u u(n) - 2g_u \frac{\mathrm{d}^2}{\mathrm{d}n^2}u(n). \tag{76}$$

We can solve this analytically to obtain an unnormalized oscillator wavefunction

$$u(n) \;=\; \mathrm{Ai}\left(\frac{n}{(2g_u)^{\frac{1}{3}}} - 2.33810\right) \qquad \text{for } n \geq 0, \tag{77}$$

$$\lambda' \;=\; -4g_u. \tag{78}$$

The constant offset 2.33810 here is intended to make the Airy function be zero for incremental $n = 0$. For $n < 0$ the oscillator states in this approximation are omitted.

7. Donor and Receiver Model with Unstable Receiver States

The reviewer noted that it was unclear from the discussion presented so far how the model connects with earlier work, and what impact it might have on cold fusion models. In our view the impact is enormous; however, it will take some further discussion beyond what we can do in this paper to make clear quite how important the result is. However, one thing that can be done, given the present result, is to consider the impact of the new model on the donor and receiver model presented previously [9].

7.1. Brief review of the donor and receiver model

The first indication that a large quantum could be fractionated with a substantial associated rate for coherent energy exchange came with the introduction of the lossy spin–boson model. With strong coupling between a set of two-level systems and an oscillator, and with loss in the vicinity of the two-level transition energy, the lossy spin–boson model predicts coherent energy exchange between the two systems under conditions of resonance. The absence of energetic particle emission commensurate with the excess energy produced in the Fleischmann–Pons experiment underscores the need for such a mechanism, and we have been optimistic now for more than a decade that lossy spin–boson models could lead to a fundamental understanding of the new physical mechanism.

However, the experiments seem to point to ^{4}He as a product, perhaps with 24 MeV energy release per atom, which seems consistent with the mass difference between two deuterons and the ^{4}He nucleus. Because of Coulomb repulsion between the two deuterons, we would expect the phonon exchange matrix element for the $D_2/^4$He transition to be very small under any reasonable set of assumptions. We could not make use of the lossy spin–boson model directly for a D_2 to ^{4}He transition. This motivated us to introduce the donor and receiver generalization of the lossy spin–boson model.

In the donor and receiver model, the donor two-level systems are assumed to be weakly coupled to the oscillator (consistent with the $D_2/^4$He transition), and the receiver two-level systems are strongly coupled with the oscillator in order to accomplish the fractionation of the large quantum as a lossy spin–boson model. The associated Hamiltonian is [9]

$$\hat{H} = \Delta E_1 \frac{\hat{S}_z^{(1)}}{\hbar} + \Delta E_2 \frac{\hat{S}_z^{(2)}}{\hbar} + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V_1 e^{-G}(\hat{a}^\dagger + \hat{a}) \frac{2\hat{S}_x^{(1)}}{\hbar} + V_2(\hat{a}^\dagger + \hat{a}) \frac{2\hat{S}_x^{(2)}}{\hbar} - i\frac{\hbar}{2}\hat{\Gamma}(E). \tag{79}$$

The analysis of the model was based on the fact that the donor coupling is very weak, as indicated by the e^{-G} Gamow factor in association with the donor coupling term in the model. As a result, we worked with basis states of the coupled receiver and oscillator model, which were available from the earlier analysis of the lossy spin–boson model. The donor coupling could then be treated simply using perturbation theory, relying on transitions between the different lossy spin–boson states of the coupled receiver and oscillator system to describe the coherent energy exchange associated with sequential resonant donor transitions.

The dynamics of the donor system that results can be described in the classical limit by

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2} m_1(t) = \frac{2}{\hbar^2} \frac{\mathrm{d}}{\mathrm{d}m_1} [V_1^{\mathrm{eff}}]^2, \tag{80}$$

where m_1 describes the donor excitation, and where V_1^{eff} is the indirect coupling matrix element for a resonant donor transition, where the coupled oscillator and receiver transition take up the donor energy. This indirect coupling matrix element was found to be

$$V_1^{\mathrm{eff}} = 2V_1\sqrt{n}\, \mathrm{e}^{-G} |\langle v_n(\phi_2)|v_{n+\Delta n_1}(\phi_2)\rangle| \sqrt{S_1^2 - m_1^2}. \tag{81}$$

Significant in this equation is that the indirect coupling matrix element is proportional to the magnitude of the overlap matrix element between two lossy spin–boson states (which is a weak function of the phase angle ϕ_2).

7.2. Subdivision in the donor and receiver model

Since some time has passed since the donor and receiver model was published, it seems helpful to update the discussion to take advantage of the pulse and amplitude approximation for the lossy spin–boson model [21]. As was noted in [9] the donor and receiver model describes a subdivision effect, so that we can think of the donor transition energy ΔE_1 as being split among many receiver transitions, each with an energy of ΔE_2, plus an offset energy that accounts for the mismatch. For this we may write

$$\Delta E_1 = \Delta N \Delta E_2 + \delta E, \tag{82}$$

where ΔN receiver excitations can be associated with a donor transition. For this case, we can make use of the pulse and amplitude approximation to estimate

$$|\langle v_n(\phi_2)|v_{n+\Delta n_1}(\phi_2)\rangle| \; = \; f\left(\frac{\delta E}{(2g_u)^{1/3}\hbar\omega_0}\right) f\left(\frac{\Delta N}{(2g_a)^{1/3}}\right), \tag{83}$$

where for the lossy spin–boson model with stable upper states for the receiver the two dimensionless coupling constants are

$$g_u \; = \; \left(\frac{V_2\sqrt{n}}{\hbar\omega_0}\right)\sqrt{S_2^2 - m_2^2} \qquad \text{(stable)}, \tag{84}$$

$$g_a \; = \; \left(\frac{V_2\sqrt{n}}{\Delta E_2}\right)\sqrt{S_2^2 - m_2^2} \qquad \text{(stable)}, \tag{85}$$

where we have indicated that these pulse and amplitude approximation parameters are for the lossy spin–boson model with stable upper states. The hindrance factor $f(y)$ is defined by

$$f(y) \; = \; \frac{\displaystyle\int_{-2.33810}^{\infty} \mathrm{Ai}(x)\mathrm{Ai}(x+y)\mathrm{d}x}{\displaystyle\int_{-2.33810}^{\infty} \mathrm{Ai}^2(x)\mathrm{d}x}. \tag{86}$$

This function is illustrated in Fig. 6.

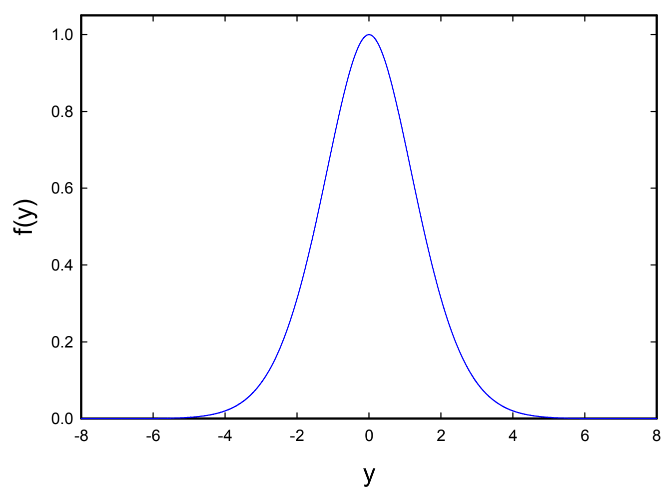

Figure 6. Plot of $f(y)$ as a function of y.

7.3. Donor and receiver model, lossy upper states

In light of the discussion of the previous sections of this paper, we are motivated to consider how the donor and receiver model would be modified were we to use receiver transitions with unstable upper states. In this case we might write the model as

$$\hat{H} = \left(\Delta E_1 \frac{\hat{S}_z^{(1)}}{\hbar}\right)_{\text{stable}} + \left(\Delta E_2 \frac{\hat{S}_z^{(2)}}{\hbar}\right)_{\text{unstable}} + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V_1 \mathrm{e}^{-G}(\hat{a}^\dagger + \hat{a})\frac{2\hat{S}_x^{(1)}}{\hbar} + V_2(\hat{a}^\dagger + \hat{a})\frac{2\hat{S}_x^{(2)}}{\hbar} - i\frac{\hbar}{2}\hat{\Gamma}(E) \tag{87}$$

to make clear that the upper state of the donor transition is stable, and the upper state of the receiver transition is unstable. Otherwise, the analysis of the model would be very similar. Since the coupling with the donor is very weak, we would compute the states of the coupled unstable receiver and oscillator model

$$E_n\Phi_n = \left\{\left(\Delta E_2 \frac{\hat{S}_z^{(2)}}{\hbar}\right)_{\text{unstable}} + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V_2(\hat{a}^\dagger + \hat{a})\frac{2\hat{S}_x^{(2)}}{\hbar} - i\frac{\hbar}{2}\hat{\Gamma}(E)\right\}\Phi_n. \tag{88}$$

Then we would make use of perturbation theory to develop an approximation for the indirect coupling matrix element. In the end, we obtain

$$V_1^{\text{eff}} = 2V_1\sqrt{n}\,\mathrm{e}^{-G}|\langle\Phi_n|\Phi_{n+\Delta n_1}\rangle|\sqrt{S_1^2 - m_1^2}. \tag{89}$$

We can make use of the pulse and amplitude approximation for the unstable case for the approximation

$$|\langle\Phi_n|\Phi_{n+\Delta n_1}\rangle| = f\left(\frac{\Delta n_1}{(2g_u)^{1/3}}\right). \tag{90}$$

In this case there is no subdivision or offset energy; the receiver transition with the unstable upper state has to accept all of the donor energy, or else no transition occurs. In this case we may write

$$g_u = \left(\frac{2V_2^2 n}{\hbar\omega_0 \Delta E_2}\right)(2S_2) \qquad (\text{unstable, two} - \text{level}). \tag{91}$$

7.4. Further generalization of the model

In the event that we have more complicated receiver systems that are not described by simple two-level systems, with many transitions in each isotope, with many different isotopes, and perhaps with different spin states of the different isotopes, then we can make use of the results in the earlier sections to write

$$|\langle\Phi_n|\Phi_{n+\Delta n_1}\rangle| = f\left(\frac{\Delta n_1}{(2g_u)^{1/3}}\right) \tag{92}$$

with

$$g_u = \sum_\beta \sum_\kappa \frac{2[V_{1\kappa}^{(\beta)}]^2 n_0 N_1^{(\beta)}}{\hbar\omega_0 E_\kappa^{(\beta)}} \qquad \text{(general unstable case).} \tag{93}$$

In this case the sum is over all receiver nuclei (κ) and all receiver transitions in each nucleus (β).

7.5. Discussion

When we found the new relativistic coupling mechanism presented in [13] our initial response was to assume that every transition in every nucleus in the lattice would contribute to the fractionation in the way described by the models outlined in this section. Since most of the coupling strength occurs for transitions with unstable upper states, the total amount of excitation is small, so the analytic approximation described in Section 6 will be applicable. In a sense this appeared to provide a simple and perhaps elegant extension of the donor and receiver model that could be adapted for the coupling of the physical system.

After we worked out the calculations outlined above, we used realistic estimates for the coupling matrix elements to estimate fractionation within the model; unfortunately, we found that essentially no fractionation occurs in this model. At first this seemed to be a crushing blow, as we had been very optimistic about the model, and it did not seem obvious what could be done to salvage the model. As we have noted at various conferences, we have analyzed a very large number of models and variants, in each case so far coming to the conclusion that the model was not consistent with experiment; consequently, it is not a rare occurrence to prove that a given model isn't right in this sense.

As mentioned in the Introduction, these negative results motivated us to seek a modification of the approach which might breathe life back into the models, which was done under the gun in preparing for ICCF17. The result of this effort was the development of a new kind of model which takes advantage of both unstable and stable transitions, and which appears to give results that have a nontrivial connection with experiment [14,15]. The new model is closely related to those under discussion in this section. We will described the new model and results obtained with it in more detail in following publications.

8. Discussion and Conclusions

We have analyzed an extension of the lossy spin–boson model where the upper state of the two-level system is unstable, assuming as before that the loss where important is taken to be infinite. The unstable upper states are taken into account in this model by restricting the available oscillator states to include only basis states above a fixed threshold, in order to prevent energy exchange from the oscillator to cause any real excitation of the two-level systems.

A key feature of the earlier lossy spin–boson model was that coherent energy exchange between the two-level systems and oscillator could occur at a rate orders of magnitude greater than expected for the (lossless) spin–boson model. Since the upper states are unstable in this model, there is no equivalent coherent energy exchange leading to real (as opposed to virtual) excitation of the two-level systems. Instead we find a simpler mixing of the degrees of freedom, and our focus has been on the ground state of the coupled system which we expect to be produced during the (adiabatic) evolution of the system as an increase in oscillator excitation results in stronger coupling between the two degrees of freedom.

In physical systems that motivate our interest in this kind of model there are present different isotopes, different spin states for the ground state of a given isotope, and in general many excited states which can be coupled to with phonon exchange. As a result we considered a generalization of the model suitable for analyzing these more complicated, but more relevant, physical systems. When many such identical nuclei are present, we find from the associated Dicke factors that the strongest transitions couple to and from ground states, which greatly simplifies the problem. We used a product solution approximation for this model, which allowed us to determine constraints for the excited state distributions and for the distribution of oscillator states. We are able to develop a useful analytic continuum model for the oscillator distribution based on this approach.

Stimulated by reviewer comments, we considered how the donor and receiver model is changed when the receiver transition has an unstable upper state. In the original donor and receiver model it was possible to subdivide the large donor quantum into many donor excitations, with an offset energy to be fractionated by the receiver; in the new version of the model subdivision is no longer since there can be no real excitation of the receiver transitions. We summarized results in terms of the equivalent classical evolution of the donor system, which depends on the strength of the receiver coupling in order to make transitions.

As discussed above a major motivation of this modeling effort has been to identify receiver transitions within the context of the donor and receiver model with sufficiently strong coupling to be able to fractionate a 24 MeV quantum in connection with the Fleischmann–Pons experiment. We had hoped that the new relativistic interaction described in [13] would lead to a large enough coupling to do the job. Since the strongest coupling with this interaction occurs for transitions with very highly excited nuclear states that are very unstable, we were motivated to carry out the analysis of the model described in this paper. However, using this new model for the receiver in the donor and receiver model does not solve the problem. There are no transitions with sufficiently strong coupling that can fractionate the 24 MeV (under conditions relevant to experiment) required to make a connection with the Fleischmann–Pons experiment. As a result, we end up with the conclusion that the basic donor and receiver model does not describe the excess heat effect in these experiments; we need a stronger model to account for the experimental results. Fortunately, such a model has been found recently [14,15], and will be discussed further in a following paper.

Appendix A. Loss Models

A reviewer has expressed concern that the inclusion of loss in the Hamiltonian has the potential to lead to difficulties, and that there exist other loss models in the literature (such as the Caldirola–Kanai model [23,24]) that might be used instead. This in our view is an important comment, and it seems appropriate to take the opportunity in this Appendix to discuss oscillator loss models briefly, and to consider loss models we are interested in.

Appendix A.1. Approaches to oscillator loss in the literature

Since the lossy oscillator constitutes the prototypical example of a dissipative system, it is no surprise that there appear a great many papers addressed to the problem. Within the general area, there are a number of specific topics that focus on aspects of the problem: such as how one quantizes a dissipative system [23,24]; simple models that are useful for analytic calculations [25,26]; general models used for modeling specific physical systems [27,28]; and advanced mathematical approaches available for analyzing such problems [29]. Our focus in the discussion here will be on the simple and more general loss models, since the other topics are not relevant to what we have presented in this paper, or in earlier papers.

Appendix A.2. Standard loss models

One can find models in the literature in which loss is taken into account coupling to a bath of oscillators; in the case of a lossy oscillator we might adopt a Hamiltonian of the form [30]

$$\hat{H} = \hbar\omega_0 \hat{a}^\dagger \hat{a} + \sum_j \hbar\omega_j \hat{b}_j^\dagger \hat{b}_j + \sum_j K_j (\hat{a} + \hat{a}^\dagger)(\hat{b}_j + \hat{b}_j^\dagger). \tag{A.1}$$

The reference oscillator in this case has frequency ω_0 and creation and annihilation operators $\hat{a}^\dagger$ and $\hat{a}$; the oscillators that make up the bath would have frequencies ω_j in the vicinity of ω_0, and we have used $\hat{b}_j^\dagger$ and $\hat{b}_j$ for the associated creation and annihilation operators. This model relies on linear coupling between the reference and bath oscillators to provide loss. A related model based on a two-level system coupled to a bath of oscillators is considered in [31].

Used less often are models where a bath of two-level systems is adopted rather than a bath of oscillators. In this case we might consider a Hamiltonian based on

$$\hat{H} \;=\; \hbar\omega_0 \hat{a}^\dagger \hat{a} + \sum_j \Delta E_j \left(\frac{\hat{s}_z}{\hbar}\right)_j + \sum_j K_j (\hat{a} + \hat{a}^\dagger) \left(\frac{2\hat{s}_x}{\hbar}\right)_j . \tag{A.2}$$

Here the bath is described using many site-dependent pseudo-spin operators $\hat{s}_x$ and $\hat{s}_z$. Note that in both cases, the basic model is explicitly Hermitian. Loss comes about from the interaction of the reference oscillator with a bath of oscillators or two-level systems that have a frequency distribution which can be chosen to match any physical loss process of interest.

The basic idea in such models is that the oscillator that is being focused on can lose energy by phonon exchange with a bath, one quantum at a time, eventually reaching thermal equilibrium with the bath. The problem simplifies if the bath is taken to be at zero temperature, in which case the oscillator eventually decays to the ground state.

Appendix A.3. Caldirola–Kanai model

The Caldirola–Kanai model is a much simpler model based on an oscillator with a dynamical mass, such as

$$\hat{H} \;=\; \frac{\hat{p}^2}{2m(t)} + \frac{1}{2}m(t)\omega_0^2 x^2 \;=\; \mathrm{e}^{-\gamma t}\frac{\hat{p}^2}{2m} + \mathrm{e}^{\gamma t}\frac{1}{2}m\omega_0^2 x^2. \tag{A.3}$$

The increasing mass in this case causes a reduction in velocity, which mimics dissipative loss in the sense that the expectation value of position satisfies

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}\langle x\rangle + \gamma \frac{\mathrm{d}}{\mathrm{d}t}\langle x\rangle \;=\; -\, m\omega_0^2 \langle x\rangle. \tag{A.4}$$

A nice feature of the model is that one can develop generalized classical states for it analytically, which makes it convenient for analyzing forced lossy oscillator models approximately. This model is widely used in the literature. There are technical issues associated with the model, as the "loss" is due to a mass increase rather than dissipation so that the uncertainty relation is not obeyed as time goes to infinity [32].

One could imagine making use of such a simple model to account for conventional oscillator loss in the models that we have studied (and based on the reviewer's comment, this may be a project to pursue in the future). Unfortunately, we would not expect it to be relevant to the loss which is responsible for the enhanced coherent energy exchange effect in the many-quantum regime that we have focused our efforts on, since it relies on a dynamical mass to mimic loss rather than modeling higher-energy loss processes (as we will discuss further below).

Appendix A.4. Spin–boson model without loss

To discuss loss in our lossy-spin boson models, the best place to start is with a spin–boson model first without loss; we write

$$\hat{H} = \hbar\omega_0 \hat{a}^\dagger \hat{a} + \Delta E \frac{\hat{S}_z}{\hbar} + V \frac{2\hat{S}_x}{\hbar}(\hat{a}^\dagger + \hat{a}). \tag{A.5}$$

In the absence of coupling ($V = 0$), then the resulting states will be pure oscillator and two level system states of the form

$$\Psi = |S, m\rangle |n\rangle. \tag{A.6}$$

When we begin to turn on the interaction, so that V is small but finite, then we would expect that the pure eigenstate will develop a small admixture of nearby states. In this case we would write approximately

$$\begin{aligned}\Psi = &\ c_0|S, m\rangle|n\rangle \\ &+ c_1|S, m-1\rangle|n-1\rangle + c_2|S, m-1\rangle|n+1\rangle + c_3|S, m+1\rangle|n-1\rangle + c_4|S, m+1\rangle|n+1\rangle.\end{aligned} \tag{A.7}$$

We see that the pure $|S, m\rangle|n\rangle$ state now will have a small admixture of states with $m \pm 1$. We would expect the state energy to be near the pure state energy

$$E_{m,n} = m\Delta E + n\hbar\omega_0. \tag{A.8}$$

For the admixed states with $m - 1$, the basis state energies are much lower than E since we assume that

$$\Delta E \gg \hbar\omega_0. \tag{A.9}$$

In the absence of loss channels this superposition presents no problem, and we view the admixed states simply as providing an off-resonant (virtual) contribution.

Appendix A.5. Spin–boson model with loss

However, things change dramatically when we augment the model with loss. Suppose we augment the spin–boson model with a bath of oscillators, consistent with

$$\hat{H} = \hbar\omega_0 \hat{a}^\dagger \hat{a} + \Delta E \frac{\hat{S}_z}{\hbar} + V \frac{2\hat{S}_x}{\hbar}(\hat{a}^\dagger + \hat{a}) + \sum_j \hbar\omega_j \hat{b}_j^\dagger \hat{b}_j + \sum_j K_j (\hat{a}^\dagger + \hat{a})(\hat{b}^\dagger + \hat{b}). \tag{A.10}$$

The idea here is that $\hat{a}$ and $\hat{a}^\dagger$ refer to the highly excited oscillator, while the oscillators described by $\hat{b}_j$ and $\hat{b}_j^\dagger$ constitute the bath.

Now, it is true that the coupling of the bath will produce oscillator loss, leading ultimately to the thermalization of the oscillator as before. But from our perspective, this isn't the most important thing that happens; more important in connection with energy exchange is the decay of the admixed states with $m - 1$. Now these admixed states have allowed coupling to decay channels in which the primary oscillator gains or loses a single additional quantum, but where a bath oscillator with energy $\hbar\omega_j$ near ΔE is excited.

Perhaps it is useful to spell this out in the case of a bath at zero temperature. If we first assume that V is small, and $K_j = 0$, then we might write the admixture above as

$$\Psi = c_0|S,m\rangle|n\rangle|\Phi_0\rangle + c_1|S,m-1\rangle|n-1\rangle|\Phi_0\rangle + c_2|S,m-1\rangle|n+1\rangle|\Phi_0\rangle + c_3|S,m+1\rangle|n-1\rangle|\Phi_0\rangle + c_4|S,m+1\rangle|n+1\rangle|\Phi_0\rangle, \quad (A.11)$$

where $|\Phi_0\rangle$ is the ground state of the bath oscillators.

If next we allow for loss, so that $K_j \neq 0$, the relevant admixture will be

$$\begin{aligned}\Psi = {} & c_0|S,m\rangle|n\rangle|\Phi_0\rangle + c_1|S,m-1\rangle|n-1\rangle|\Phi_0\rangle + c_2|S,m-1\rangle|n+1\rangle|\Phi_0\rangle \\ & + c_3|S,m+1\rangle|n-1\rangle|\Phi_0\rangle + c_4|S,m+1\rangle|n+1\rangle|\Phi_0\rangle \\ & + \sum_j d_{0,j}|S,m\rangle|n+1\rangle\, \hat{b}^\dagger|\Phi_0\rangle + \sum_j e_{0,j}|S,m\rangle|n-1\rangle\, \hat{b}^\dagger|\Phi_0\rangle \\ & + \sum_j d_{1,j}|S,m-1\rangle|n\rangle\, \hat{b}^\dagger|\Phi_0\rangle + \sum_j e_{1,j}|S,m-1\rangle|n-2\rangle\, \hat{b}^\dagger|\Phi_0\rangle \\ & + \sum_j d_{2,j}|S,m-1\rangle|n+2\rangle\, \hat{b}^\dagger|\Phi_0\rangle + \sum_j e_{2,j}|S,m-1\rangle|n\rangle\, \hat{b}^\dagger|\Phi_0\rangle \\ & + \sum_j d_{3,j}|S,m+1\rangle|n\rangle\, \hat{b}^\dagger|\Phi_0\rangle + \sum_j e_{3,j}|S,m+1\rangle|n-2\rangle\, \hat{b}^\dagger|\Phi_0\rangle \\ & + \sum_j d_{4,j}|S,m+1\rangle|n+2\rangle\, \hat{b}^\dagger|\Phi_0\rangle + \sum_j e_{4,j}|S,m+1\rangle|n\rangle\, \hat{b}^\dagger|\Phi_0\rangle. \end{aligned} \quad (A.12)$$

The admixed states with $d_{0,j}$ and $e_{0,j}$ coefficients are involved with conventional oscillator loss. The associated process is consistent with normal thermalization of the oscillator (in this case to zero temperature ultimately), and consistent with oscillator loss models in which there are no two-level systems.

However, the admixed states with $d_{1,j}$ and $e_{1,j}$ coefficients, and also with $d_{2,j}$ and $e_{2,j}$ coefficients, are new and special. The reason for this is that the system has now coupled to states in which the two-level system energy ΔE is given to the bath, under conditions where the bath has resonant states with energy $\hbar\omega_j = \Delta E$ and $\hbar\omega_j = \Delta E \pm 2\hbar\omega_0$. Because of the mixing with these states, we might expect incoherent decay process to occur, which we could evaluate using the Golden Rule decay formula.

The same is not true for the admixed states with $d_{3,j}$ and $e_{3,j}$ coefficients, and also with $d_{4,j}$ and $e_{4,j}$ coefficients. Although they appear in the admixture, there are now no resonant states in the bath because we have $m+1$ states with an extra two-level system excited. The resonance conditions would have to be $\hbar\omega_j = -\Delta E$ and $\hbar\omega_j = -\Delta E \pm 2\hbar\omega_0$, which is impossible since the bath oscillator frequencies are positive.

The enhancement of the coherent energy exchange rate in the multi-quantum regime comes about because loss channels are present generally for basis states with energies below E, and restricted for basis states with energies above E. This breaks the interference effect in which contributions from the two groups of states destructively interfere in connection with coherent energy exchange in the multi-quantum regime.

Note that in this kind of model we are making use of oscillator models to account for energetic transitions that may have nothing to do with any oscillator. For example, such an energetic loss process might involve atom ejection, electron ejection, or a nuclear decay. However, since loss impacts these models in essentially the same way independent of the particular loss channel, we would expect that any bath model which describes loss channels in the relevant energy regime will behave similarly.

Appendix A.6. Sectors

Loss can be analyzed by making use of sectors in connection with infinite-order Brillouin–Wigner theory; although the approach is not so widely used in condensed matter problems these days. It is possible to make clear how this works within the context of our modeling effort. For simplicity it will be convenient to assume that the bath starts in the ground state. In this case it will be useful to make use of two sectors: one in which the bath remains in the ground state; and one in which the bath has at least one excitation. We can divide the wavefunction into two sector wavefunctions

$$\Psi = \Psi_A + \Psi_B, \tag{A.13}$$

where Ψ_A will denote the sector with the zero-temperature bath, and Ψ_B will contain all states with an excited bath. We might denote time independent Schrödinger equation for Ψ as

$$E\Psi = \hat{H}\Psi. \tag{A.14}$$

This can be rewritten in terms of the two different sectors as

$$E\Psi_A + E\Psi_B = \hat{H}_{AA}\Psi_A + \hat{H}_{AB}\Psi_B + \hat{H}_{BA}\Psi_A + \hat{H}_{BB}\Psi_B, \tag{A.15}$$

where the Hamiltonian is split into pieces which preserve sector, and which change sector. It is possible to separate this into two sector equations

$$\begin{aligned} E\Psi_A &= \hat{H}_{AA}\Psi_A + \hat{H}_{AB}\Psi_B, \\ E\Psi_B &= \hat{H}_{BA}\Psi_A + \hat{H}_{BB}\Psi_B. \end{aligned} \tag{A.16}$$

The overall problem is still explicitly Hermitian.

If we were to focus only on sector A, then $\hat{H}_{AA}$ would appear to us to be Hermitian relative to that sector, but $\hat{H}_{AB}$ would seem to us not to be Hermitian since it couples to a different sector. So, within the context of an explicitly Hermitian formulation, we can have a situation in which part of a Hermitian Hamiltonian is going to act as if it is not Hermitian within sector A.

In the infinite-order Brillouin–Wigner formulation, we can write the sector B wavefunction in terms of the source from sector A as

$$\Psi_B = \left(E - \hat{H}_{BB}\right)^{-1} \hat{H}_{BA}\Psi_A. \tag{A.17}$$

This can be used to write the sector A part of the eigenvalue equation as

$$E\Psi_A = \hat{H}_{AA}\Psi_A + \hat{H}_{AB}\left(E - \hat{H}_{BB}\right)^{-1} \hat{H}_{BA}\Psi_A. \tag{A.18}$$

Even at this point the problem remains Hermitian, but in this form it is written so that we have isolated sector A, and taken into account the effect of sector B in a complicated infinite-order Brillouin–Wigner operator. Using this approach, we might think of the sector A wavefunction as being governed by a complicated second-order Hamiltonian of the form

$$\hat{H}_A = \hat{H}_{AA} + \hat{H}_{AB}\left(E - \hat{H}_{BB}\right)^{-1}\hat{H}_{BA}. \tag{A.19}$$

Such a model is still Hermitian, since it has not discarded any part of the original Hermitian problem, and the original eigenvalues can in principle be recovered (although there are technical issues in doing so).

When the decay is exponential, this infinite-order Brillouin–Wigner formulation can be used to obtain a Golden Rule estimate for the decay rate directly

$$\gamma = -\frac{2}{\hbar}\mathrm{Im}\left\{\hat{H}_{AB}\left(E - \hat{H}_{BB}\right)^{-1}\hat{H}_{BA}\right\}. \tag{A.20}$$

It is possible to take advantage of this to write for the sector A Hamiltonian

$$\hat{H}_A \rightarrow \hat{H}_{AA} - \frac{i\hbar\hat{\Gamma}(E)}{2}. \tag{A.21}$$

One could argue at this point that the resulting model is now not Hermitian, and then argue that all results obtained from such a model are suspect. Another might argue that since sector A experiences loss which goes to sector B, no loss of probability appears in the overall model so the overall problem remains Hermitian. For some applications having an explicit loss operator capable of giving an accurate estimate for the sector loss might be considered to be an advantage.

Since the enhanced coherent energy exchange rate in the lossy spin–boson model comes about by eliminating the destructive interference associated with the different virtual states (as described above), there are only minor differences between the results if an accurate loss model is used compared to simply making the loss infinite whenever a loss channel is open. When the decay rate for a state becomes infinitely fast, then it accumulates no occupation probability, so it is the same as if the state were not included in the first place. We have found it convenient to model loss then by assuming infinitely fast decay, and eliminating the associated states. The exclusion of states done in this way leads to a sector A Hamiltonian that is Hermitian, since there is no net decay from the sector if all loss channels are infinitely fast (as long as some accessible stable states remain).

Appendix A.7. Discussion

In view of the discussion above, we conclude that there are two distinct oscillator loss mechanisms which are important in the lossy spin–boson models. One loss mechanism is conventional, in which the oscillator couples a single oscillator energy quantum $\hbar\omega_0$ at a time to roughly resonant bath modes. This loss is connected to the fast thermalization of high frequency oscillator modes that is observed experimentally, and is modeled in the literature using standard approaches. It would be possible to imagine making use of the Caldirola–Kanai model (or some other simple model) in connection with this basic type of oscillator loss.

The other loss mechanism is unconventional, in which the oscillator couples an energetic quantum with energy near the two-level system transition energy ΔE to energetic modes in the bath via single phonon exchange. In this case, it is perhaps more useful to think about the two-level systems and oscillator as a coupled system which couples the bath (instead of the oscillator alone coupling to the bath). With such a view, it is natural to expect the coupling to involve energy exchange $\hbar\omega_0$ and also ΔE, since the coupled two-level system and oscillator has excitations in the vicinity of both energies. It is this phonon loss mechanism that is responsible for the enhancement in the coherent multiphonon energy exchange which is the hallmark of the lossy spin–boson models that we have studied. We would not expect the Caldirola–Kanai model, or other simple oscillator loss models, to result in an enhancement of the coherent energy exchange rate.

The underlying model for our work is one in which the physical loss mechanisms can be thought of as represented by a bath of oscillators over a large energy range. Then, within the framework of the infinite-order Brillouin–Wigner formalism, the energetic decay processes responsible for the elimination of destructive interference effects are replaced formally by a loss operator (resulting in a sector Hamiltonian that as written is non-Hermitian). Finally, for quantitative estimates we take the limit of infinitely fast loss, which is equivalent to the exclusion of the states which experience the loss. In the end, the sector model is Hermitian, since there is no net decay from the excluded states.

For more accurate modeling in the future where quantitative estimates for state distributions and loss rates in the presence of finite loss will be important, then we will have to work with a explicitly non-Hermitian sector Hamiltonian if we retain an infinite-order Brillouin–Wigner formalism. At that time some effort would be required to be sure that the results are reliable. But for the detailed calculations so far Hermitian sector Hamiltonians have been used for all results over the past several years.

References

[1] P.L. Hagelstein and I.U. Chaudhary, Energy exchange in the lossy spin-boson model, *J. Condensed Matter Nucl. Sci.* **5** (2011) 52.

[2] P.L. Hagelstein and I.U. Chaudhary, Second-order formulation and scaling in the lossy spin-boson model, *J. Condensed Matter Nucl. Sci.* **5** (2011) 87.

[3] P.L. Hagelstein and I.U. Chaudhary, Local approximation for the lossy spin–boson model, *J. Condensed Matter Nucl. Sci.* **5** (2011) 102.

[4] P.L. Hagelstein and I.U. Chaudhary, Coherent energy exchange in the strong coupling limit of the lossy spin-boson model, *J. Condensed Matter Nucl. Sci.* **5** (2011) 116.

[5] M. Fleischmann, S. Pons and M. Hawkins, *J. Electroanal. Chem.* **201** (1989) 301; errata **263** (1990) 187.

[6] M. Fleischmann, S. Pons, M.W. Anderson, L.J. Li and M. Hawkins, *J. Electroanal. Chem.* **287** (1990) 293.

[7] P.L. Hagelstein, M. C. H. McKubre, D. J. Nagel, T. A. Chubb and R. J. Hekman, *Proc. ICCF11* (2004) 23.

[8] P.L. Hagelstein, *Naturwissenschaften* **97** (2010) 345.

[9] P.L. Hagelstein and I.U. Chaudhary, Generalization of the lossy spin-boson model to donor and receiver systems, *J. Condensed Matter Nucl. Sci.* **5** (2011) 140.

[10] P.L. Hagelstein, Bird's eye view of phonon models for excess heat in the Fleischmann–Pons experiment, *J. Condensed Matter Nucl. Sci.* **6** (2011) 169.

[11] D. Letts, D. Cravens and P.L. Hagelstein, Dual laser stimulation and optical phonons in palladium deuteride, *Low-Energy Nuclear Reactions and New Energy Technologies Sourcebook*, American Chemical Society, Washington DC, Volume 2, 2009, p. 81.

[12] P.L. Hagelstein, D. Letts and D. Cravens, *J. Condensed Matter Nucl. Sci.* **3** (2010) 59.

[13] P.L. Hagelstein and I.U. Chaudhary, Including nuclear degrees of freedom in a lattice Hamiltonian, *J. Condensed Matter Nucl. Sci.* **7** (2012) 35.

[14] P.L. Hagelstein and I.U. Chaudhary, A model for collimated X-ray emission in the Karabut experiment, *Proc. ICCF17 J. Condensed Matter Nucl. Sci.* **6** (2012) 217.

[15] P.L. Hagelstein and I.U. Chaudhary, Models for excess heat in PdD and NiH, *Proc. ICCF17* (in press).

[16] F. Bloch and A. Siegert, *Phys. Rev.* **57** (1940) 522.

[17] J. Shirley, *Phys. Rev.* **138** (1965) B979.

[18] C. Cohen-Tannoudji, J. Dupont-Roc and C. Fabre, *J. Phys. B: Atomic Mol. Phys.* **6** (1973) L214.

[19] P.L. Hagelstein and I.U. Chaudhary, Dynamics in the case of coupled degenerate states, *J. Condensed Matter Nucl. Sci.* **5** (2011) 72.

[20] P.L. Hagelstein and I.U. Chaudhary, *J. Phys. B: Atomic Mol. Phys.* **41** (2008) 035601.

[21] P.L. Hagelstein and I.U. Chaudhary, Pulse and amplitude approximation for the lossy spin–boson model, *J. Condensed Matter Nucl. Sci.* **9** (2012) 30.
[22] P.L. Hagelstein and I.U. Chaudhary, Coupling between a deuteron and a lattice, *J. Condensed Matter Nucl. Sci.* **9** (2012) 50.
[23] P. Caldirola, Forze non conservative nella meccanica quantistica, *Il Nuovo Cimento* **18** (1941) 393.
[24] E. Kanai, On the quantization of the dissipative systems, *Prog. Theoret. Phys.* **3** (1948) 440.
[25] V.V. Dodonov and V.I. Man'ko, Coherent states and the resonance of a quantum damped oscillator, *Phys. Rev. A* **20** (1979) 550.
[26] F. Riew, Mechanics with fractional derivatives, *Phys. Rev. E* **55** (1997) 3581.
[27] P.S. Riseborough, P.Hänggi and U. Weiss, Exact results for a damped quantum-mechanical oscillator, *Phys. Rev. A* **31** (1985) 471.
[28] M. Thorwart and P. Hänggi, Decoherence and dissapation during a quantum XOR operation, *Phys. Rev. A* **65** (2001) 012309.
[29] M. Blasone and P. Jizba, Quantum mechanics of the damped harmonic oscillator, *Can. J. Phys.* **80** (2002) 645.
[30] K. Lindenberg and B.J. West, Statistical properties of quantum systems: the linear oscillator, *Phys. Rev. A* **30** (1984) 568.
[31] R. Silbey and R.A. Harris, Variational calculation of the dynamics of a two level system interacting with a bath, *J. Chem. Phys.* **80** (1984) 2615.
[32] C.-I. Um, K.-H. Yeon and T.F. George, The quantum damped harmonic oscillator, *Physics Reports* **362** (2002) 63.