

Research Article

# Energy Exchange In The Lossy Spin-Boson Model

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## Abstract

Excess heat in the Fleischmann–Pons experiment is observed without commensurate energetic particles, which is inconsistent with known nuclear reactions. Any proposed model must address this at the outset. At present, we recognize are two general approaches to the problem: mechanisms which transfer the reaction energy directly to a condensed matter mode, which requires the fractionation of a large quantum into many small quanta; mechanisms in which the large energy quantum is converted to kinetic energy over a very large number of neighboring nuclei. We have focused on the first approach, and we have found a model that seems to accomplish this. Here we introduce the model, which we call the “lossy spin-boson model”. We find that energy exchange in the lossless spin-boson model is hindered due to destructive interference effects. Augmenting the model with loss removes the destructive interference, and we use perturbation theory on a specific example for illustration. Feshbach projection operators and the Brillouin–Wigner formalism which we adopt to describe loss in the model are reviewed in Appendix A.

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## 1. Introduction

Over the past several years, we have been thinking about models relevant to excess heat in the Fleischmann–Pons experiment [1,2]. In these experiments, a prodigious amount of energy is observed, and  $^4\text{He}$  is seen in the gas phase in amounts correlated quantitatively with the energy produced [3–5]; essentially no energetic particles are observed correlated with the energy [6]. This latter aspect of the experiments has drawn our attention over the years as being of particular importance.

To understand why, we consider briefly the situation in a “normal” nuclear reaction. In a Rutherford picture, the incident nuclei might be taken to be as classical particles, and similarly the product nuclei might also be thought about

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classically. Both energy and momentum are conserved in such a picture, so that the basic kinematic relations for the final state products can be determined from a knowledge of the reaction energy and initial state momenta and energies. A consequence of this is that the reaction energy in the case of deuteron-deuteron fusion ends up as kinetic energy of the proton and triton, or neutron and  $^3\text{He}$  nucleus, in the final state. In the quantum mechanical description of the reaction, energy and momentum conservation similarly requires that the reaction energy appears as kinetic energy of the product nuclei.

In the Fleischmann–Pons experiment, there are no energetic particles commensurate with the energy produced. In light of the comments above, one might reasonably conclude that no nuclear reactions take place. But if so, then what is the source of the energy, and where does the  $^4\text{He}$  come from? In the early days following the initial announcement of 1989, arguments were put forth that the energy was a result of chemical reactions, or experimental error [7]. The arguments against chemical reactions include the absence of observable commensurate chemical reaction products, and that the amount of energy is much greater in some cases than a chemical level of energy associated with all the atoms of the cathode and electrolyte. The primary argument against experimental error at this time is that the effect has been reported in a very large number of experiments [8,9], where different calorimeters and different types of calorimetry have been used.

So, what can be the resolution? Energy at the nuclear level is seen in many experiments,  $^4\text{He}$  seems to be present as a product that is correlated with the energy produced, and yet there are no commensurate energetic particles as would be required in a Rutherford-type picture of a nuclear reaction. We are by now sure that energy is produced as an experimental fact; we are pretty sure that  $^4\text{He}$  is produced as an experimental fact; and we are sure that commensurate energetic particles are not present as an experimental fact. In essence, some new kind of reaction seems to be taking place in a way inconsistent with the Rutherford picture of how a nuclear reaction is supposed to work.

If so, then where does the energy go? From experiment, we measure it as thermal energy; but whatever intermediate forms the energy might appear in prior to thermalization so far is hidden.

There is indirect evidence in the Letts two-laser experiment that the energy goes into optical phonon modes (in this experiment in particular) stimulated at the beat frequency, since excess heat persists after the lasers are turned off [10,11]. The theoretical question as to how such a thing might happen is at present an open one.

In our efforts to understand the effect, we have been interested in the possibility of the fractionation of a large energy quantum into a very large number of smaller energy quanta [12]. The basic idea is that the initial nuclear reaction energy will be measured in units of MeV, while the characteristic energy of an optical phonon mode as an oscillator is measured in units of meV. This suggests that perhaps the large nuclear quanta is somehow being split up into on the order of  $10^8$  smaller energy quanta. If a mechanism exists to accomplish this, then we might be able to understand how excess heat is produced in the Fleischmann–Pons experiment.

Although this approach has seemed perhaps the most plausible one to us, there is no precedent, and the majority of our colleagues favor various other approaches. Among the schemes that are presently being considered there are aneutronic fusion schemes in which the reaction products are hidden [13]; there is the approach of Kim wherein the reaction energy is somehow partitioned as kinetic energy among a very large number of deuterium atoms [14]; and there is the general approach of Widom and coworkers [15] where the reaction products are again hidden. There have been a great many proposals for reaction mechanisms put forth over the years, some of which are based on aneutronic mechanisms that are otherwise largely conventional. The intuition associated with such proposals is that a reasonably unreactive energetic product (such as an alpha) would be able to slow down and thermalize inside the cathode so as to be “hidden”. The measurements described in [6] can be interpreted, based on direct calculations of the associated secondary yields, as placing an upper limit on the energy of a product alpha to be conservatively less than 20 keV. In essence, energetic alphas produced at the watt level cannot be “hidden” in a PdD cathode. Since  $^4\text{He}$  appears as a primary product of the new process, this upper limit on the kinetic energy is a very severe constraint, and rules out the majority of proposed mechanisms involving  $^4\text{He}$  that have been proposed. We do not think that it is possible to “hide”

commensurate energetic reaction products [6], and we do not understand what kind of direct physical mechanism is capable of distributing a large reaction energy (for example, 24 MeV) over more than  $10^4$  deuterium atoms as proposed by Kim.

In the case of direct coupling of the reaction energy into condensed matter modes, there is no real precedent for such an effect. The results outlined here follow from our recent studies of basic models involving two-level systems and harmonic oscillators that began around 2000. The thinking behind this kind of model was that the two-level system would stand in for two nuclear states with an MeV energy difference, and the oscillator would stand in for a vibrational mode of the lattice. The question of interest was whether there was any way to exchange energy coherently between the two systems; our earlier results (from the previous decade) suggested that that it would be extremely difficult to couple a relevant number of quanta directly from two-level systems to the oscillator. Computations of coupled systems were done under a variety of assumptions: weak coupling, strong coupling, linear interactions and nonlinear interactions. In these computations, energy exchange seemed to be restricted to less than 100 oscillator quanta at a time.

The situation changed qualitatively when we recognized that energy exchange in these systems was inhibited by massive destructive interference, which could be removed in lossy systems. We presented calculations at ICCF9 in 2002 which showed the basic effect [16]. We used a quantum flow formulation at that time, and carried out calculations which showed that coherence was maintained under conditions where up to  $10^4$  oscillator quanta were exchanged. This result seemed to us to be quite significant, since it showed that coherent energy exchange could occur between two-level systems and an oscillator with incommensurate quanta. Following the initial presentation at ICCF9, there seems to have been little outside interest in the result.

Subsequently, we devoted significant effort to better understand the new models. In the absence of loss, the early models that we had explored are equivalent to the spin-boson model which has received considerable attention in the literature [17–19] (but much less attention in regard to the issue of energy exchange in the multiphoton regime). We developed a rotation that allowed us to treat coherent multiphoton energy exchange in the spin-boson model in the same way as one would analyze linear coupling [20,21]. In this approach, we were able to isolate dressed states which were nearly degenerate, but “distant” with regard to the linear coupling of the model, and estimate the indirect coupling between them analytically.

The lossy problem is much harder, and to date we have not identified an analogous rotation that would allow a similar treatment of the problem. However, we have over the years applied a number of different techniques which have allowed us to characterize the model and establish important limits. In this manuscript, we begin our discussion of the lossy spin-boson model. We start with a brief consideration of the spin-boson model, and then augment the spin-boson model with loss using a Brillouin–Wigner loss operator. Since the projection operators of Feshbach and infinite-order Brillouin–Wigner theory were used more often in decades past, we provide a brief review in Appendix A. Finally, we demonstrate explicitly that indirect coupling between distant states is hindered by destructive interference in the spin-boson model using perturbation theory (which provides for the cleanest demonstration of the effect), and that the indirect coupling is not hindered in the lossy model (by considering the limit where the loss becomes infinite for states with an energy excess).

In the course of the review process, a great many criticisms were put forth by a reviewer, who wanted them to be addressed systematically in this paper. The issues of interest cover a lot of topics, and we chose to focus our attention on a subset of the problems so that papers of manageable size might result. Nevertheless, some of our colleagues thought that the associated discussion might be useful, so we have added selections of the reviewer’s criticisms along with some responses in Appendix B.

## 2. Spin-boson Model Augmented with Loss

The spin-boson model is a popular model which includes two-level systems and an oscillator with linear coupling. Earlier versions of the model were investigated in association with NMR research about 70 years ago [17], and extended to problems involving atoms and electromagnetic fields about 35 years ago [18]. From our perspective, this Hamiltonian is interesting because it describes the most basic interaction between two fundamental quantum models (two-level systems and a harmonic oscillator), and more particularly, it sheds light on coherent energy exchange between the two systems when the two-level transition energy is a large multiple of the oscillator energy.

### 2.1. Spin-boson model

The Hamiltonian for the spin-boson model is [19]

$$\hat{H} = \frac{\Delta E}{\hbar} \hat{S}_z + \hbar \omega_0 \hat{a}^\dagger \hat{a} + V(\hat{a}^\dagger + \hat{a}) \frac{2\hat{S}_x}{\hbar}. \quad (1)$$

In this model,  $\Delta E$  is the transition energy of the two-level systems,  $\hbar \omega_0$  is the characteristic energy of the oscillator, and  $V$  is a coupling strength.

Our intuition might lead us to anticipate that energy exchange between the two systems should occur when the transition energy of the two-level systems is matched to the characteristic energy of the oscillator. This is so when the coupling is weak. If the coupling is stronger, and if there is a near resonance between the two-level transition energy and three oscillator quanta, then once again energy can be exchanged coherently. The reason that this works is that the system is able to maintain coherence over the time that several interactions occur, so that a nonlinear response is possible even though the coupling is linear.

We have explored energy exchange in this model in the multiphoton regime, with much larger numbers of oscillator quanta exchanged for a single two-level system quantum [20,21]. Unfortunately, coherent energy exchange in the multiphoton regime is a weak effect that requires very precise level matching. Coherent energy exchange at finite rates in this model is possible under moderately strong coupling for up to about 50 quanta at a time; for the exchange of even more quanta, the constraints on the degree of resonance becomes prohibitive, and the energy exchange rates become very small [23].

### 2.2. Lossy spin-boson model

If we augment the spin-boson model with loss, things change qualitatively in regard to energy exchange in the multiphoton limit as we will show later on in this paper. We write such a model as [22–24]

$$\hat{H} = \frac{\Delta E}{\hbar} \hat{S}_z + \hbar \omega_0 \hat{a}^\dagger \hat{a} + V(\hat{a}^\dagger + \hat{a}) \frac{2\hat{S}_x}{\hbar} - i \frac{\hbar \hat{\Gamma}(E)}{2}. \quad (2)$$

The loss term in this model is an operator that comes about from an infinite-order Brillouin–Wigner model as discussed in Appendix A. As written, this loss term is completely general, but we are thinking of it as being referenced to the oscillator so as to provide strong loss at energies in the vicinity of the two-level system transition energy.

In regard to the Fleischmann–Pons model, the oscillator here stands in for an optical phonon mode, so that  $\hbar \omega_0$  is tens of meV. The transition energy  $\Delta E$  for the two-level system is assumed to be associated with nuclear energy levels, on the order of MeV. The thought regarding the loss term here is that loss channels for the phonon mode open if the system has available several MeV of energy through the disintegration of nuclei that make up the phonon mode.

### 3. Indirect Coupling in Perturbation Theory

We have argued previously that the introduction of loss into the model leads to an enormous enhancement of indirect coupling between distant states that are nearly degenerate. To understand this better, if we consider a product basis state of the form

$$|S, m\rangle|n\rangle,$$

then the Hamiltonian leads to coupling between neighboring states of the form

$$|S, m \pm 1\rangle|n \pm 1\rangle.$$

If the two-level transition energy  $\Delta E$  were matched to the characteristic energy  $\hbar\omega_0$  of the oscillator, then this direct coupling would lead to coherent energy exchange on or near resonance. However, in the multiphoton regime of the model, we require the exchange of more oscillator quanta to bring the system back into energy balance. As a result, the closest states which are nearly degenerate are the distant states

$$|S, m - 1\rangle|n + \Delta n\rangle, |S, m + 1\rangle|n - \Delta n\rangle,$$

where  $\Delta n$  is the number of oscillator quanta exchanged. There is no direct coupling between  $|S, m\rangle|n\rangle$  and these states; when the coupling constant is sufficiently large there can be a weak indirect coupling. It is this weak indirect coupling that we are interested in here.

#### 3.1. Indirect coupling in perturbation theory with no loss

Indirect coupling is very weak in the lossless spin-boson model because of destructive interference. We can see this most readily in a perturbation theory calculation. As an example, we consider indirect coupling in the case where 5 oscillator quanta are exchanged between the nearly degenerate basis states  $|S, m\rangle|n\rangle$  and  $|S, m + 1\rangle|n - 5\rangle$ .

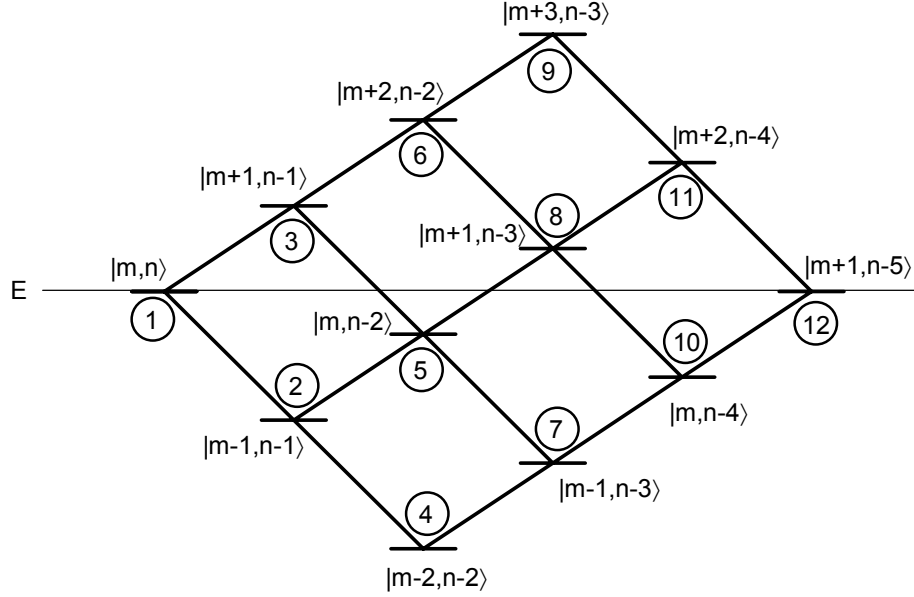
We can develop the perturbative result that we are interested in starting from a finite basis expansion of the form

$$\begin{aligned} \Psi &= \sum_j c_j \Phi_j \\ &= c_1 |S, m\rangle|n\rangle + c_2 |S, m - 1\rangle|n - 1\rangle + c_3 |S, m + 1\rangle|n - 1\rangle + c_4 |S, m - 2\rangle|n - 2\rangle \\ &\quad + c_5 |S, m\rangle|n - 2\rangle + c_6 |S, m + 2\rangle|n - 2\rangle + c_7 |S, m - 1\rangle|n - 3\rangle + c_8 |S, m + 1\rangle|n - 3\rangle \\ &\quad + c_9 |S, m + 3\rangle|n - 3\rangle + c_{10} |S, m\rangle|n - 4\rangle + c_{11} |S, m + 2\rangle|n - 4\rangle + c_{12} |S, m + 1\rangle|n - 5\rangle. \end{aligned} \quad (3)$$

The numbering here corresponds to the state definitions indicated in Fig. 1.

The finite basis equations for the coefficients can be written as

$$\begin{aligned} Ec_1 &= m\Delta E + n\hbar\omega_0 + V_{1,2}c_2 + V_{1,3}c_3, \\ Ec_2 &= (m - 1)\Delta E + (n - 1)\hbar\omega_0 + V_{2,1}c_1 + V_{2,4}c_4 + V_{2,5}c_5, \\ &\vdots \end{aligned}$$



**Figure 1.** Basis states involved at lowest order in perturbation theory for indirect coupling with the exchange of five oscillator quanta.

$$Ec_{12} = (m+1)\Delta E + (n-5)\hbar\omega_0 + V_{12,10}c_{10} + V_{12,11}c_{11}. \quad (4)$$

To develop these equations, we require the diagonal and off-diagonal matrix elements of the spin-boson Hamiltonian. The nonzero matrix elements are

$$\begin{aligned} \langle n | \langle S, m | \hat{H} | S, m \rangle | n \rangle &= M\Delta E + n\hbar\omega_0, \\ \langle n+1 | \langle S, m+1 | \hat{H} | S, m \rangle | n \rangle &= V\sqrt{n+1}\sqrt{(S-m)(S+m+1)}, \\ \langle n-1 | \langle S, m+1 | \hat{H} | S, m \rangle | n \rangle &= V\sqrt{n}\sqrt{(S-m)(S+m+1)}, \\ \langle n+1 | \langle S, m-1 | \hat{H} | S, m \rangle | n \rangle &= V\sqrt{n+1}\sqrt{(S+m)(S-m+1)}, \\ \langle n-1 | \langle S, m-1 | \hat{H} | S, m \rangle | n \rangle &= V\sqrt{n}\sqrt{(S+m)(S-m+1)}. \end{aligned} \quad (5)$$

Since our focus is on the indirect coupling between  $|S, m\rangle|n\rangle$  and  $|S, m+1\rangle|n-5\rangle$ , we can keep the corresponding coefficients  $c_1$  and  $c_{12}$ , and eliminate the others algebraically. This leads to coupled equations of the form

$$\begin{aligned} Ec_1 &= m\Delta E + n\hbar\omega_0 + \Sigma_1(E)c_1 + V_{1,12}(E)c_{12}, \\ Ec_{12} &= (m+1)\Delta E + (n-5)\hbar\omega_0 + \Sigma_{12}(E)c_{12} + V_{12,1}(E)c_1. \end{aligned} \quad (6)$$

Here  $\Sigma_1(E)$  and  $\Sigma_{12}(E)$  are self-energies, and  $V_{1,12}(E)$  and  $V_{12,1}(E)$  are indirect coupling coefficients. The lowest-order contribution to the indirect coupling coefficient  $V_{1,12}(E)$  is obtained by summing over the different paths

$$V_{1,12}(E) = \frac{V_{1,2}V_{2,4}V_{4,7}V_{7,10}V_{10,12}}{(E - H_2)(E - H_4)(E - H_7)(E - H_{10})} + \frac{V_{1,2}V_{2,5}V_{5,7}V_{7,10}V_{10,12}}{(E - H_2)(E - H_5)(E - H_7)(E - H_{10})} + \dots, \quad (7)$$

where  $H_j$  is the diagonal matrix element of basis state  $j$ . There are 10 terms altogether in this summation. Assuming resonance  $H_1 = H_{12}$ , this sum can be evaluated in closed form to yield

$$V_{1,12}(E) = \frac{625}{64} \frac{V^5 \sqrt{n(n-1)(n-2)(n-3)(n-4)}}{\Delta E^4} \sqrt{(S-m)(S+m+1)}. \quad (8)$$

### 3.2. Destructive interference

The indirect coupling term in the lossless case is very small due to destructive interference. This can be seen from a comparison of the summed indirect coupling coefficient  $V_{1,12}(E)$  with the first term of the series of Eq. (7). We may write

$$\frac{V_{1,2}V_{2,4}V_{4,7}V_{7,10}V_{10,12}}{(E - H_2)(E - H_4)(E - H_7)(E - H_{10})} = \frac{625}{64} \frac{V^5 \sqrt{n(n-1)(n-2)(n-3)(n-4)}}{\Delta E^4} \sqrt{(S-m)(S+m+1)} \frac{(S+m)(S+m-1)(S-m+1)(S-m+2)}{36} \quad (9)$$

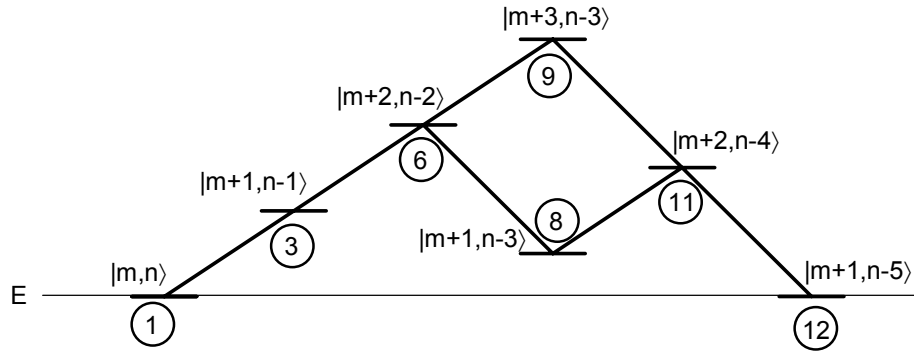
If the number of two-level systems is large, then  $S$  will be large. This term can be larger than the sum by a factor that can be on the order of  $S^4/36$ , which gives an indication of how effective destructive interference is for this case.

### 3.3. Indirect coupling in the lossy model

In the augmented version of the problem with loss, the destructive interference can be removed if the loss terms are sufficiently large. We can begin to see this in perturbation theory by using a finite basis approximation with the same basis states as we used above. This time, the first three expansion coefficients satisfy coupled equations of the form

$$\begin{aligned} Ec_1 &= H_1c_1 + V_{1,2}c_2 + V_{1,3}c_3, \\ Ec_2 &= (H_2 - i\hbar\Gamma_2/2)c_2 + V_{2,1}c_1 + V_{2,4}c_4 + V_{2,5}c_5, \\ Ec_3 &= H_3c_3 + V_{3,1}c_1 + V_{3,5}c_5 + V_{3,6}c_6. \end{aligned} \quad (10)$$

Intermediate states (such as  $\Phi_2$ ) which have energies less than  $E$  have decay modes available in this model, since the system can decay to such states if energy is lost through the decay processes modeled by the loss operator. Other states (such as  $\Phi_3$ ) which are in energy deficit as intermediate states do not have analogous decay modes available, and are



**Figure 2.** Basis states involved at lowest order in perturbation theory for indirect coupling with the exchange of five oscillator quanta in the limit of infinite loss.

free of loss in this model. States that are resonant (such as  $\Phi_1$  and  $\Phi_{12}$ ) are assumed to be stable against zero-energy decay processes.

The expression one obtains in such a model for the indirect coupling coefficient is very complicated, and not particularly enlightening. As we are interested in the issue of destructive interference between different pathways, we can eliminate the destructive interference completely by considering a model in which the loss terms are much greater than other terms that appear in the denominator. In the limit that the associated loss is infinite, then only pathways which include intermediate states in energy deficit contribute (see Fig. 2). In this limit we sum to obtain

$$V_{1,12}(E) \rightarrow \left[ V_{1,12}(E) \right]_{\Gamma_j=0} F(S, M) \quad (\Gamma_j \rightarrow \infty), \quad (11)$$

where the  $\Gamma_j = 0$  result is given in Eq. (8), and where the factor  $F(S, M)$  evaluates to

$$F(S, M) = \frac{(S - m - 1)(S + m + 2)(7S^2 + 7S - 7m^2 - 23m - 18)}{36}. \quad (12)$$

We see from this calculation that destructive interference greatly reduces the indirect coupling between distant resonant states in the original Dicke model, leading to a much larger indirect coupling matrix element.

#### 4. Discussion and Conclusions

We are interested in the possibility that a large nuclear quantum of energy can be fractionated and exchanged with a condensed matter mode. To investigate this possibility, we have focused on the simplest basic physics model which exhibits coherent energy exchange between relevant quantum systems – the spin-boson model augmented with loss. This model is interesting in that it describes coherent energy exchange at usefully large rates under conditions where a large two-level system energy quantum is divided into a great many small energy quanta. In our most recent studies with more realistic and more sophisticated models, the indirect coupling appears to be strong enough to allow for coherent energy transfer at rates relevant to excess heat production in the Fleischmann–Pons experiment. This provides us with motivation for pursuing this model and its generalizations.

We presented the spin-boson Hamiltonian along with the generalization to the lossy version of the problem. Although one can find in the literature versions of the spin-boson model augmented with loss, these generalizations of the model

are usually concerned with oscillator loss near the characteristic frequency of the oscillator (so that the oscillator is damped). These models are qualitatively different from the one we have given, since we are interested in oscillator loss generally at higher frequency which produces a different physical effect.

We used perturbation theory to argue that indirect coupling between distant nearly degenerate states is hindered by destructive interference in the spin-boson model (without loss), and that this destructive interference is removed in the lossy problem when the loss is large. This establishes the most important property of the new model in a clean and obvious way, that it allows for very much enhanced indirect coupling.

The new model is moderately complicated despite its deceptively simple sector Hamiltonian, so it seemed helpful to provide a review of projection operators and infinite-order Brillouin–Wigner theory in Appendix A. This seems appropriate given that the version of the model that we have been studying is expressed using this formalism, and such a review may be helpful for our colleagues in the field.

It remains to establish the scaling of the new model with respect to its dimensionless coupling constant, to develop a formalism that allows us to understand it using a formalism based on indirectly coupled nearly degenerate states, and to establish formulas for the indirect coupling in the limit that a very large number of quanta are exchanged. These issues will be dealt with in following manuscripts.

## Appendix A. Second-order Loss Models

From the earliest days of quantum mechanics, the inclusion of loss into models has been of interest. Over the years a variety of different approaches have been introduced of varying degrees of complexity. In our work, we have made use of an approach which is sometimes called infinite order Brillouin–Wigner theory. While this approach was at one time a very standard approach, in recent years it has not been as popular as other methods. This motivates us to provide a brief review in this section.

### Appendix A.1. Sectors

We consider a simple example of a system which has a set of states that we can identify readily in which the system resides prior to decaying, which we will refer to as  $\{\Psi_a\}$ ; and a second set of states that we can readily associate with the system after it has decayed, which we will refer to as  $\{\Psi_b\}$ . By identifying these two distinct sets of states in this way, we divide the associated Hilbert space, or Fock space, into two different sectors.

We assume that there is a Hamiltonian  $\hat{H}$  that is valid for all of the states of the system, and that eigenstates satisfy

$$E\Psi = \hat{H}\Psi. \quad (\text{A.1})$$

We can divide the wavefunction  $\Psi$  into different pieces corresponding to the different sectors. One way to do this is to use projection operators ( $\hat{P}$  and  $\hat{Q}$ ) [25] so that

$$\Psi_a = \hat{P}\Psi, \quad \Psi_b = \hat{Q}\Psi. \quad (\text{A.2})$$

The first projection operator  $\hat{P}$  separates out the part of the wavefunction  $\Psi$  that belongs in the first sector; and the second operator  $\hat{Q}$  does the same for the second sector. If we assume that there are only two sectors of interest, then we may write

$$\hat{P} + \hat{Q} = 1. \quad (\text{A.3})$$

### Appendix A.2. Sector equations

We can take advantage of these operators to isolate the parts of the Hamiltonian  $\hat{H}$  which refer to the different sectors, which leads to sector equations. We may write

$$E \hat{P} \Psi = \hat{P}(\hat{H} \Psi) \quad (\text{A.4})$$

to project into the  $a$  sector. Since

$$\hat{H} \Psi = \hat{H}(\hat{P} + \hat{Q}) \Psi, \quad (\text{A.5})$$

we can expand and write

$$E \Psi_a = (\hat{P} \hat{H} \hat{P}) \Psi_a + (\hat{P} \hat{H} \hat{Q}) \Psi_b, \quad (\text{A.6})$$

where we have used the identities

$$\hat{P}^2 = \hat{P}, \quad \hat{Q}^2 = \hat{Q}. \quad (\text{A.7})$$

Similarly, we may write

$$E \Psi_b = (\hat{Q} \hat{H} \hat{Q}) \Psi_b + (\hat{Q} \hat{H} \hat{P}) \Psi_a. \quad (\text{A.8})$$

### Appendix A.3. Sector Hamiltonian with loss

The projection operators have allowed us to separate cleanly the problem into two pieces, but now we have a situation in which the coupling terms between the different sectors are not Hermitian. This is important, and also useful, since when there is loss we expect to lose probability from the first sector.

It is possible to eliminate  $\Psi_b$  algebraically from the  $a$  sector equation, which will produce a second-order Hamiltonian for the  $a$  sector that includes simple loss effects. We can do this by first writing

$$\Psi_b = [E - (\hat{Q} \hat{H} \hat{Q})]^{-1} (\hat{Q} \hat{H} \hat{P}) \Psi_a, \quad (\text{A.9})$$

which allows us to obtain the second-order equation for the  $a$  sector

$$E \Psi_a = (\hat{P} \hat{H} \hat{P}) \Psi_a + (\hat{P} \hat{H} \hat{Q}) [E - (\hat{Q} \hat{H} \hat{Q})]^{-1} (\hat{Q} \hat{H} \hat{P}) \Psi_a. \quad (\text{A.10})$$

The second-order term in general is complex; the real part is an effective potential, and the imaginary part gives rise to loss. We might express this as

$$\hat{\Sigma}(E) = \text{Re} \left[ (\hat{P} \hat{H} \hat{Q}) [E - (\hat{Q} \hat{H} \hat{Q})]^{-1} (\hat{Q} \hat{H} \hat{P}) \right], \quad (\text{A.11})$$

$$-\frac{\hbar \Gamma(E)}{2} = \text{Im} \left[ (\hat{P} \hat{H} \hat{Q}) [E - (\hat{Q} \hat{H} \hat{Q})]^{-1} (\hat{Q} \hat{H} \hat{P}) \right]. \quad (\text{A.12})$$

Our sector Hamiltonian  $\hat{H}_a$  might be written as

$$\hat{H}_a(E) = (\hat{P} \hat{H} \hat{P}) + \hat{\Sigma}(E) - i \frac{\hbar \hat{\Gamma}(E)}{2}. \quad (\text{A.13})$$

#### Appendix A.4. Connection with Golden Rule decay

On the one hand, this might seem to be a rather strange Hamiltonian, since it depends explicitly on the energy eigenvalue  $E$ , and since it is not Hermitian if loss is present. On the other hand, this formalism can be very useful since it includes decay very simply, and since the loss term in the Hamiltonian is consistent with the Golden Rule decay rate [26]

$$\gamma = \frac{2}{\hbar} \left\langle \text{Im} \left[ (\hat{P} \hat{H} \hat{Q}) [E - (\hat{Q} \hat{H} \hat{Q})]^{-1} (\hat{Q} \hat{H} \hat{P}) \right] \right\rangle = \frac{2\pi}{\hbar} |\langle \Psi_b | \hat{Q} \hat{H} \hat{P} | \Psi_a \rangle|^2 \rho(E_f). \quad (\text{A.14})$$

The eigenvalues of this second-order  $a$  sector Hamiltonian are complex, so that if we solve

$$E \psi_a = \hat{H}_a(E) \psi_a \quad (\text{A.15})$$

then we can find the associated decay rate for a state from

$$\gamma = -\frac{2}{\hbar} \text{Im}\{E\}. \quad (\text{A.16})$$

## Appendix B. Reviewer Comments and Responses

The first reviewer for this and the following paper had a great many criticisms. To address all of the issues raised by the reviewer in the main text did not seem to be a reasonable thing to do, as it would have greatly extended the length and complexity of the paper. Our approach has been to recognize at the outset that the problem of interest is complicated, and made up of a great many pieces, so that we might make progress best by focusing on a small number of issues at a time.

Some of our colleagues thought that the associated discussion was helpful. As a result, we have decided to include a subset of the comments (which we have in some cases restated or revised), along with some associated comments. In some cases these comments were those in our response, in other cases we have summarized comments from our response, and in yet others we have provided different and hopefully better comments.

### Appendix B.1. On the need for many step-wise levels

As  ${}^4\text{He}^*(E_x)$  nuclear excited states are known to be limited for  $E_x = 23.8, 23.35, 23.04, 21.04, 21.01, 20.21$  MeV after the  $d+d$  reaction, possible electromagnetic transitions from the  $E_x = 23.8$  MeV state are limited for step-wise emitting photon quanta of 0.45, 0.31, 0.03, 0.8, 20.21, 0.76, 0.83, 2.7, 3.6 and 23.8 MeV; even neglecting the spin-parity selection rule (as spin-parity of 23.8 MeV state is unknown). These are all easily measurable gamma-rays by an HPGe detector, but they are never observed. Therefore the proposed transition with very small quanta (30 meV;  $3 \times 10^{-2}$  eV) stepwise is not possible as the corresponding  ${}^4\text{He}$  excited states do not exist step-wise.

To avoid this difficulty, one must show that very fine (30 meV step, for instance) new levels with defined spin-parities are possible by the third interaction field. This is not done in the present paper (once Talbot Chubb proposed such an idea, but has never proved it by his theory).

This comment is the one most relevant to the technical content of our papers. From our perspective, we are grateful that the reviewer has spelled out this argument, since it very much helps to clarify a very pervasive misunderstanding about the models we have studied.

In the conventional spin-boson model, significant rates for coherent energy exchange occur when several tens of oscillator quanta make up a single two-level system quantum. There are no intermediate states available for a step-wise excitation as the reviewer has mentioned in the associated model. I note that there are reports of experimental results in which this kind of energy exchange has been observed directly, in agreement with the theoretical models.

I agree that many people have intuition that suggests that the only way to exchange a lot of quanta with an oscillator is by having a lot of equi-spaced levels. Many years ago, I started from the same place, and at one time I used this very argument to dismiss the possibility of energy exchange with a low energy oscillator. However, the argument is simply wrong. One does not need equi-spaced levels in order to exchange a large number of quanta with an oscillator. In the case of the spin-boson model, this was known at least as early as 1965 when Shirley's paper [18] appeared; there is now a sub-field that focuses on this regime (which is called the multi-photon regime in the literature). In our paper, we show that this is true for the lossy version of the spin-boson model, and obtain an analytic result in the simplifying case when the loss is infinite.

#### Appendix B.2. The Coulomb barrier problem

*At the end of reading through the paper, the reviewer noticed a big problem. The paper does not describe about how to overcome the Coulomb barrier, for two deuterons to make their relative distance very close for inducing nuclear fusion reaction by the strong interaction under mutual very short range interaction (nuclear confinement) potential. Did they solve the problem in their past works? Probably not. The paper does not use the relative distance between two deuterons as key variable for solving wave functions of quantum-mechanical equations with defined Hamiltonians. As a consequence, the solution, how very close d-d pairs are realized in the assumed nuclear-lattice-phonon coupled-particle confining potential, is not shown.*

The first two papers introduce the lossy spin-boson model; make clear that destructive interference is the reason that coherent energy exchange is hindered in the normal spin-boson model; demonstrates a strongly enhanced coherent energy exchange effect under conditions where a modest number of oscillator quanta are exchanged; and then presents a tool with which we can analyze this kind of model. However, the reviewer wants to see a solution to the problem of tunneling through the Coulomb barrier, a discussion of excited states of  $^4\text{He}$ , confining potentials, and many other topics as well. All of this is well out of the scope of the papers under consideration. The inclusion of material to deal with these issues would greatly lengthen the papers, and make them incomprehensible to readers.

Nevertheless, the Coulomb barrier appears in more sophisticated versions of the model, and we have worried a great deal about the associated tunneling contribution to the matrix element. We note that in a coherent reaction theory, the reaction rate can be linear in the matrix element, where the reaction rate goes as the square of the matrix element in incoherent reaction models [so the Gamow factor can come in as  $\exp(-G)$  rather than as  $\exp(-2G)$ ]. This provides a natural way within the theory to obtain a dramatic enhancement of the associated reaction rate while taking into account tunneling through the Coulomb barrier.

In early versions of our models from many years ago, we made use of the molecular  $\text{D}_2$  potential to estimate the Gamow factor numerically. In recent years, we have found that the coherent reaction rate that results from such an assumption ends up being much too small to account for experimental observations. The inclusion of screening effects at the level of  $U_e$  near 100 eV greatly improves the agreement between model predictions and experiment. Such values for the screening energy  $U_e$  are close to those estimated in the Thomas-Fermi approximation by Czerski and coworkers [27], who obtain  $U_e = 133.8$  eV for  $\text{PdD}_{0.2}$ .

### Appendix B.3. Derivation of nuclear-phonon coupling

*The imagined “direct coupling mechanism between nuclear excited states and solid-state lattice optical phonons ( $10^4$  quanta)” is not plausible in view of EM-nuclear-force coupling. First we need to treat nuclear-EM coupling and next we may treat succeeding coupling between EM-waves (virtual photons) and lattice optical phonons. The present paper does not consider such necessary path; hence, the proposed spin-boson model is in question.*

We have not included a derivation of the model in these papers in order to restrict our focus to a manageable set of issues.

So far, there does not seem to exist a theory in the literature that allows such computations to be done in the case of a  $D_2$  to  $^4\text{He}$  transition. We have in fact worked out a theory along the lines of what the reviewer suggests, and presented it at ICCF14 [28]. This approach can be used for coupling between molecular  $D_2$  states and the  $^4\text{He}$  ground state.

For transitions involving ground state nuclei as might be relevant to receiver systems in more complicated versions of the model, we are exploring phonon exchange associated with nuclear transitions that are dipole and quadrupole coupled to atomic electrons in the context of three- or more-level models.

The inclusion of associated discussions and derivations is out of the scope of the present set of papers. If we included such material here, the resulting modified papers would become very long, and would be difficult to read.

### Appendix B.4. Magnitude of the coupling matrix element

*The formalism of the “spin-boson model” seem OK, but only if the DIRECT and significant-magnitude coupling between the nuclear strong interaction (to MeV level intermediate compound state) and the lattice optical phonons exists. However, this is surely in doubt. Extending the elegant mathematics looks off-centered. The authors should explain the supporting logic of it. If not, the treated transition between two levels seems to be forbidden by itself; and it is nonsense to develop the mathematics associated with the coherent dynamics between the two levels.*

We are not aware of a way to develop the effects of interest by pursuing transitions to intermediate compound states as seems to be required by the reviewer. We are also not aware of any general result that argues that phonon exchange is forbidden in association with transitions between different nuclear states.

We are able to derive the relevant phonon exchange matrix elements (as noted above), and the associated transitions are not forbidden as proposed by the reviewer.

Implicit in the reviewer’s comment seems to be the notion that the reviewer would be able to tell if the coupling matrix element were sufficiently large to be relevant. This seems unlikely. From our perspective, one needs to analyze the models in order to figure out how large the coupling matrix elements need to be. It is only relatively recently that we have been able to establish such criteria for simplified versions of our models.

### Appendix B.5. Adoption of a solution prior to solving the problem

*It seems that they assumed that a d-d nuclear admixture or  $^4\text{He}^*$  (probably its excited energy 23.8 MeV, or something else) intermediate excited state happened to appear abruptly (or a priori) without any proper reasons of substantially shown confining mechanisms. They assume superposition of two deuteron’s wave functions are 100% both in the complex EM (electro-magnetic field) potential for many particles (palladium atoms, deuterons, electrons, lattice phonons, virtual photons for EM interaction) and in the strong interaction confining potential. Such an initial assumption is, unfortunately, common to other “theories” as Y. Kim’s BECNF. Isn’t the adoption of a solution before solving the*

*problem a forbidden methodology in theory? Anyway, they started with the desired condition that a close deuteron–deuteron pair or  ${}^4\text{He}^*$  (probably its excited energy 23.8 MeV) with very long life time (also unimaginable in nuclear physics) exists in the assumed nuclear-lattice-phonon coupled particle confining potential. The reviewer requires for the authors to explain how such a priori conditions can have consistent capability (proper logics in physics) to deduce proper solutions of the F-P excess heat problem or phenomenon if existing.*

From our perspective, the biggest theoretical issue that must be addressed in response to the Fleischmann–Pons excess heat effect is coherent energy exchange under conditions where a large quantum is fractionated into a very large number of smaller quanta. As a result, we have studied very general models involving two-level systems and an oscillator, seeking a basic understanding of this new coherent energy exchange mechanism. This is the subject of the first two papers, and the three that follow.

If we had found that these general models were not capable of the coherent energy exchange effect of interest, then there would be no reason to pursue the identification of specific states and coupling mechanisms. So, the big question of interest initially is whether the effect can be demonstrated under any conditions or set of assumptions. Once this has been done, then possible reactions scenarios can be analyzed using results from the more general models.

In any event, the reviewer seems to want to focus on the specific states and pathways first, before developing the associated coherent dynamics models. From our perspective, had we been analyzing a much simpler incoherent reaction mechanism, then we would use the Golden Rule to evaluate the reaction rates, so that all that would be needed would be a specification of the initial, intermediate, and final state reaction channels (much as the reviewer would like to see specified here). However, coherent reaction dynamics are much more difficult to analyze. There is no simple general formula analogous to the Golden Rule for coherent reaction dynamics. For the idealized lossy spin-boson model under discussion in our papers, it has taken us five papers to develop tools and results sufficient to associate the model reaction rate to the relevant matrix elements. We need all of these results, and more, in order to begin to understand how coherent reactions involving specific reaction pathways might work.

#### Appendix B.6. Impossibility of describing $d + d +$ particle reactions

*The present paper never treats the nuclear excited state after the deuteron–deuteron reaction; the spin, parity, and isospin, with excited energy value, after “some external force interaction” (lattice optical phonons are out of this scope, in principle). Therefore the “spin-boson model” of the paper has no capability to tell the final state interaction of the  $(d + d) +$  “something” reaction going out to the  ${}^4\text{He}$  ground state only, if at all, killing known break-up channels ( $p+t$  and  $n+{}^3\text{He}$ ). Thus whole story becomes a desired scenario, a-priori chosen, in imaginary thought.*

The reviewer appears to assume here that an additional particle is needed, as in  $d + d +$  “something”. We know that in vacuum,  ${}^4\text{He}$  when it occurs (with low probability) is accompanied by a 23.85 MeV gamma. Energy and momentum conservation in vacuum requires an additional particle, so that if another particle is present then it could result in another final state channel with  ${}^4\text{He}$  and no gamma.

However, if we accept that experiments have been done where excess heat is observed, and where no significant neutron emission is correlated with the excess heat (with an upper limit near 0.01 n/J) [6], then we are forced to conclude that no reaction of this kind is going to be consistent with experiment. Let us consider systematically the possibilities in the case that  $d + d +$  something is in the input channel, and  ${}^4\text{He} +$  something is in the output channel, with a 24 MeV reaction energy:

- ${}^4\text{He} + \text{Pd}$  (an example where the alpha energy is maximized), with the alpha particle ending up with about 23 MeV. Although fast alphas are not penetrating, they cause  $\alpha(d,n+p)\alpha$  deuteron break-up reactions with a

high yield, with fast neutrons that are penetrating. We calculated an expected yield of  $10^7$  n/J, which is nine orders of magnitude above the neutron per unit energy upper limit from experiment.

- $^4\text{He} + d$  (since there are deuterons in the system), so that the alpha particle ends up with about 8 MeV. We would expect about  $10^4$  n/J from the same alpha-induced deuteron break up reaction, which is now six orders of magnitude above experiment. However, the deuteron will have 16 MeV, which would make dd-fusion neutrons with a yield of just under  $10^8$  n/J, which is a bit less than 10 orders of magnitude above the upper limit from experiment.
- $^4\text{He} + p$ , so that we get the minimum alpha particle recoil for any nucleus, and the alpha ends up with 4.8 MeV. The number of secondary neutrons produced as a result of primary collisions between the alphas and deuterons in the lattice now is reduced to about 200 n/J, which is about four orders of magnitude above the experimental limit. The energetic protons in this case would cause deuteron break up reactions with a yield near  $10^7$  n/J, which is nine orders of magnitude above the experimental limit.
- $^4\text{He} + e$ , which gives close to the minimum alpha recoil for any single particle, and the alpha ends up with about 76 keV. Now the secondary neutron emission due to the alphas is down to 10 n/J, only three orders of magnitude above experiment. However, penetrating 24 MeV electrons produced at the watt level would again constitute a significant health hazard for any experimentalists nearby. For an experimentalist within a meter of an experiment producing a watt of 24 MeV betas, the radiation dose would be on the order of 1 rem/s (assuming a 10 cm range) which would be lethal in about 1 min.
- $^4\text{He} + \gamma$ , again giving 76 keV recoil energy for the alpha, and again 10 n/J which is again three orders of magnitude above experiment. Penetrating 24 MeV gammas at the watt level would be a major health hazard for any human beings in the general vicinity. As in the case of fast electrons, 24 MeV gammas at the watt level would be lethal for an exposure of about 1 min at a meter distance.
- $^4\text{He} + \text{neutrino}$  (as advocated by Li), also gives 76 keV recoil energy for the alpha, so we would expect three orders of magnitude more neutrons than the experimental upper limit. The neutrinos in this case are not a health hazard, and we would not know from direct measurements if they were there. However, most of the reaction energy would go into the neutrinos, so that the observed reaction  $Q$ -value would be about 76 keV, which differs from the experimental value by about 300.

We can conclude from this that no reaction scheme based on  $d + d + \text{particle}$  is capable of being consistent with the experimental upper limit on neutron emission in association with energy production, with the observed  $Q$ -value (energy produced per  $^4\text{He}$  observed), and with the absence of lethal amounts of electrons, gammas or other light particles.

#### Appendix B.7. Impossibility of killing off 3+1 channels

... Therefore the “spin-boson model” of the paper has no capability to tell the final state interaction of the  $(d + d) + \text{“something”}$  reaction going out to the  $^4\text{He}$  ground state only, if at all, killing known break-up channels ( $p + t$  and  $n + ^3\text{He}$ ) ...

The reviewer seems to require some particle and associated mechanism to “kill off” the  $n + ^3\text{He}$  and  $p + t$  exit channels, and faults the two-level system description as not being able to deal with this. If we focus on the two-level system part of the problem alone (since the optical phonon modes are associated with the oscillator), then there is no ability to describe in detail the microscopic physics that the reviewer believes is required in the final state to “suppress” the primary deuteron–deuteron fusion channels, and hence favor  $^4\text{He}$ .

In our view, there is no way to suppress the fast  $n + ^3\text{He}$  or  $p+t$  decay channels; with, or without, an additional particle present. When two deuterons approach each other sufficiently close for conventional fusion reactions to occur,

the 2+2 to 3+1 reactions proceed about as fast as the laws of physics allow, and there is not much that is going to stop them from happening. The reviewer seems to suggest that these channels need to be killed off somehow, perhaps through the introduction of a third body. Now, if one were to pursue such an approach, and if one were to start from the conventional fusion branching ratios, then one would need to kill off the 3+1 channels by about  $11 + 7 = 18$  orders of magnitude relative to the  $^4\text{He}$  channel to be consistent with experiment (11 orders of magnitude for the observed branching ratio, and 7 since the  $^4\text{He}$  branch occurs at a rate much slower than the 3+1 channels). From our perspective, this is simply impossible.

Our approach has been instead to focus on coherent dynamics mechanisms, which work differently. So, instead of trying to change the existing incoherent reaction pathways (which we don't think can be changed significantly anyway), our view is to focus on new coherent mechanisms that might be able to happen in addition to the existing incoherent reactions. In the end, to determine relative reaction rates, one need compare a coherent rate to an incoherent rate. There seems to be no fundamental problem in having a coherent rate be orders of magnitude larger than an incoherent rate.

We have pursued more sophisticated models in which there are more sets of two-level systems. In these models, one set of two-level systems is responsible for converting a large quantum into many small quanta. Since matrix elements between the  $\text{D}_2$  and  $^4\text{He}$  system is hindered by Coulomb repulsion, the associated coupling is very small, which means that there is no way to exchange significant energy with the lattice in the simple lossy spin-boson model. In these more sophisticated models, the energy from the  $\text{D}_2/^4\text{He}$  system is transferred to other transitions, which then convert it to phonon mode excitation. In this case, all that we require is a single phonon exchange to convert molecular  $\text{D}_2$  to  $^4\text{He}$ . We can compute the incoherent reaction rate for decay to 3+1 channels on the same footing as for the coherent reaction pathway, and we can find conditions under which the coherent pathway is many orders of magnitude greater.

#### Appendix B.8. Neutron detection not done with calorimetry

*The authors mixed up various measurements for independent items under different conditions to conclude as if all the measured data took place simultaneously in one experiment.*

In our Naturwissenschaften paper (Ref. [6]), we cited five experiments in which neutron detection was operating when excess heat bursts were seen.

- In the case of the Oak Ridge result (Scott et al. [29]), a graph showing the level of neutron emission during the time at which excess heat was seen is presented in Fig. 6 of a preliminary version of their conference proceeding paper for ICCF1 [30].
- In the case of the OSU result (Klein et al. [31]), one can see plotted temperature excursions and neutron emission levels on the same graphs as a function of time in Figs. 2–4 in their AIP conference proceedings paper.
- In the *J. Fusion Energy* paper of Wolf et al. [32], one finds written: “The correlation of the results on neutrons, tritium, and heat production has proved to be negative. Obvious heat generation (ignition) by a Srinivasan cell was not accompanied by neutron emission, and none of these cells show tritium production despite an indication of 5–15% excess heat.”
- In the ICCF3 paper by Takahashi et al. [33], results for excess heat measurements and neutron measurements using the L-H current protocol are given in Fig. 7.
- In the ICCF4 paper by Gozzi et al. [34], Fig. 9 shows excess heat results for cell #10, and Fig. 12 shows neutron counts for various neutron detectors for the same acquisition period, where detector #1 is reasonably close to cell #10.

In these references, experiments were discussed in which neutron detection ran monitoring cells that produced

excess power. In all of these cases, one of the points of the experiments reported was to determine whether the neutron emission was correlated with excess heat production. There are additional experiments that have been done in which excess heat was reported also with neutron detection operational; however, only in the references we used were we able to develop quantitative estimates in terms of source neutrons per unit energy. From the information reported in the papers cited in Ref. [6], it seems clear that neutron measurements were done on cells that produced excess heat, when they produced excess heat, in contrast to the claim of the reviewer.

#### Appendix B.9. Absence of energetic particles not yet proven

*An experimental proof showing noncorrelation of energetic out-going particles and claimed (not reproducible) excess heat in Fleischmann-Pons type electrolysis experiments is not clear at all. Accurate experimental works accurately measuring time-dependent correlations between excess heat, helium, alpha-particles with their energies, neutrons with their energies, gamma-rays, X-rays, and low energy photons has not been given. Some experiments measured once-through excess heat without accurate nuclear radiation detection; others measured nuclear radiation somehow accurately, but the calorimetry was poor, or else there was no calorimetry. Therefore the argument in Ref. [6], which is the starting assumption of paper is not certain, or wrong.*

*The very restricted view of choosing only one scenario out of Takahashi's four scenarios ([3]) is in question. The true status is that we do not have enough experimental evidences showing on-line correlations between excess heat (irreproducible), neutrons (unknown energy), alpha-particles or helium (unknown energy), gamma-rays (unknown energy), X-rays (unknown energy), EUV, EV, visible light, etc. data. Nobody has made successfully such experiments. Due to the irreproducibility of Fleischmann-Pons excess heat experiments, we do not have definite logics to make such a narrow-focus conclusion as done by Hagelstein [6]. It is premature and dangerous.*

I will be the first to agree with the reviewer that more experimental results are needed generally in the field. The controversy that has plagued the field has resulted in funding difficulties, so that a great many experiments that we would like to be done have not managed to get done. As a result, in our considerations today we are forced to deal with the experimental data set that is available today, imperfect as it may be. If new experiments are done in the future, we can revisit the associated issues.

Perhaps the most significant issue in response to what the reviewer has written concerns a misconception that you need to diagnose for all kinds of emissions ("helium, alpha-particles with their energies, neutrons with their energies, gamma-rays, X-rays, low energy photons") in order to be able to say something about whether there are energetic particles in the exit channel. In this, the reviewer is simply incorrect. The point of the discussion of Ref. [6] (and also of related papers [35–37] that appeared last year in the CMNS journal) is that if you think  $^4\text{He}$  is a reaction product, then you can say something about how much energy it has, since energetic alphas make primary or secondary neutrons very efficiently in PdD.

You do not need to detect the alphas directly, or measure their energies, for the purpose of this argument. If there are no commensurate neutrons produced, then you can be sure that there are not a corresponding number of energetic alphas as determined by the yield functions given in [6] (and in the other papers published in the CMNS journal). If the neutrons aren't there, you do not need to measure gamma-rays or X-rays to answer the question of whether there are a lot of fast alphas – there aren't, or else you would have known from the neutron measurements.

The reviewer argues that inaccurate calorimetry impacts the issue of whether commensurate energetic alphas are present. Now, let us suppose that the calorimetric errors are as large as a factor of 2 (which is substantially larger than the claimed accuracy in the different papers used in [6]). In this case, we might take the excess power to be less by 2x, which would increase the upper limits for neutrons per Joule by 2x. So, an upper limit in this case might go from 0.01 to 0.02 n/J. This does not help very much since from the examples given above, we would need on the order of 9 or

10 orders of magnitude more in order to have consistency between the number of neutrons expected and the number of neutrons seen for the different proposed incoherent reaction mechanisms with a two-body exit channel (that are not fatal to the experimentalists, and have roughly the right ratio between energy and amount of helium).

Results from reproducible experiments are preferred. But the reviewer seems to be suggesting that observations taken in the case of experiments where the reproducibility of the excess heat effect is less than 100% should be ignored. From our perspective, the issue as to whether there are energetic alphas involves a very large numbers of neutrons, so that having five published reports of observations of excess heat bursts, where each of which reports neutron emission at levels more than a factor of a million below what would be seen if energetic alphas were present in commensurate amounts, is very significant.

Both the reviewer, and the authors, would like for the associated heat bursts in these experiments to be reproducible, and even better if the measurements were more accurate. However, for us, these and other papers provide good confirmation that large numbers of neutrons simply aren't there; while this reviewer rejects all of the measurements (of these five papers, as well as all other papers – of which there are at least five more reporting similar results, but without giving sufficient numbers so that a value of neutrons per Joule could be extracted) that give similar results. Moreover, this reviewer seems unwilling to accept any relevant experiment until the excess heat effect is completely reproducible, until the calorimetry is very accurate, until the neutrons are measured with energy resolution at the same time, until energetic alphas are measured at the same time, and until also gammas, X-rays, EUV, and optical photons are measured as well. This reviewer has exceptionally high standards in what measurements will be accepted. But in our view, this matter is settled already with orders of magnitude to spare in the difference between the level of neutron emission observed, and that needed for energetic alphas to be present.

I do not think that such conclusions are either premature, dangerous, or narrowly focused. The real issue here is that energetic alphas in amounts commensurate with the energy observed produce secondary neutrons with a surprisingly high yield, so that if they were there it would be very easy to tell. They simply aren't there.

#### Appendix B.10. Chemical origin of energy in the Letts experiment

*The excess energy observed by Letts et al. [10,11] has no evidence that it is nuclear reaction energy, but may be chemical energy. So, the possibility of chemical reactions should be considered as candidate explanation of the observed results.*

The original argument that the excess energy in the Fleischmann–Pons experiment is nuclear was based on the very large amount of energy observed, combined with the relative absence of observed chemical products. A similar argument can be made for the two-laser experiment. In run 669Z, Letts observed 118 kJ of excess energy produced in about 36 h following initiation by two-laser stimulation. The cathode in these experiments have a volume of about 0.0012 cm<sup>3</sup>, which corresponds to about  $7.2 \times 10^{19}$  Pd atoms. The observed energy per Pd atom is then over 9300 eV. As in the Fleischmann–Pons experiment, there were no observed commensurate chemical reaction products.

The energy produced in the two-laser experiments used to develop the spectrum of Ref. [11] was less (closer to 10 kJ). The corresponding energy per Pd atom in this case is less (about 800 eV/Pd atom), but still well outside of what can be accounted for by chemistry.

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