Modeling Excess Heat in the Fleischmann-Pons Experiment

Project Staff
Peter L. Hagelstein, Irfan Chaudhary, Michael McKubre, Francis Tanzella, Jennifer Novosad, James Gimlett, Dennis Letts, and Dennis Cravens

Introduction

The notion of local energy and momentum conservation in a nuclear reaction provide a foundation of nuclear physics. Classical models for energy and momentum conservation were used as a way to understand nuclear scattering experiments in the time of Rutherford [1]. Energy released in a nuclear reaction is expressed through energetic particles, as consistent with all such reactions studied in nuclear physics, and also as presented in introductory sections of modern texts on the subject [2].

In the Fleischmann-Pons experiment, observations of excess heat production are not accompanied by observations of commensurate energetic nuclear particles [3]. The energy produced in these experiments is orders of magnitude greater than can be associated with chemical or solid-state processes. In the case of heavy water experiments, ^{4}He has been observed as a reaction product in the gas stream in amounts proportional to the energy produced (see the discussion in [4]). From experiment, a reaction Q has been determined experimentally to be 24 MeV to within about 10%. This is consistent with some kind of new physical process in which deuterons react to make ^{4}He, with the reaction energy going into low-energy channels.

There are no observations of nuclear reactions reported in the nuclear physics literature since the beginning of the field in the late 19th century that at present are understood to work in this way. Most nuclear physicists regard the possibility of coupling reaction energy at the MeV level to low energy degrees of freedom as impossible. The absence of evidence supporting the existence of such mechanisms is previous experimental work is consistent, from this perspective, with the impossibility of such an effect.

Such a view is supported by a simple semi-classical picture of a typical fusion process. Two deuterons must approach to within about 10 fermis in order for a nucleon to be transferred, and once this occurs, the reaction products (n+^{3}He or p+t) move away from each other with a relative velocity that is about 10% of the speed of light. Hence, there is simply no time for communication with neighboring nuclei in a condensed matter phase (this argument is discussed in [5]). One would expect intuitively that it should take a much longer time in order to develop some kind of dynamics that would involve low-energy electronic or lattice degrees of freedom. Moreover, even if such reaction channels existed, these channels would have to compete with the much faster kinetic exit channels, which seems on the face of it to be unlikely.

Because of arguments such as these, there has been a reluctance on the part of the nuclear physics community to consider, much less accept, the experimental observations of excess heat production that have accumulated over the years. Since 1989, it has been easier to accept the notion that the experiments are somehow in error, or perhaps not reproducible, or to question the credibility of those doing the experiments; than to acknowledge the challenge these results present to a foundation of nuclear physics. Quite simply, if local energy and momentum conservation holds in nuclear reactions, then the great many observations of excess heat in

Fleischmann-Pons are somehow in error. On the other hand if the excess heat effect is not in error, then local energy and momentum conservation need not hold, and a foundation of nuclear physics will require revision. Models seeking to account for excess heat production require a more general foundation.

Excitation transfer

It is possible for an atomic-scale system to violate local energy and momentum conservation through a mechanism termed excitation transfer. Dipole-dipole coupling between distant resonant systems can allow the excitation at one site to be transferred to another site. The classic demonstration of excitation transfer within a protein was reported by Bücher and Kaspers in 1947 [6]. Förster put forth in 1948 a model for resonant excitation transfer between identical fluorophores [7]. Resonance excitation transfer is currently an active area of study in biophysics [8].

In an excitation transfer process, global energy and momentum are conserved; but local energy and momentum conservation is violated at both interacting sites. The energy lost at one site appears at the other.

Excitation transfer in these applications is mediated by the Coulomb interaction; however, one would expect similar effects to occur through coupling to other degrees of freedom. Our focus in recent years has been on excitation transfer involving indirect phonon exchange as an indirect coupling mechanism. A de-excitation at one site with phonon exchange can be coupled to an excitation at another site, provided that both sites exchange phonons with a common phonon mode. This process has not been the focus of similar active study in the case of atomic or molecular excitations, although one can find mention of it in the literature [9].

In the case of excess heat production in the Fleischmann-Pons experiment, we consider D_2 at one site (inside the host metal lattice in the vicinity of a vacancy) to constitute the upper state of an equivalent two-level system, and ^{4}He to constitute the lower state [10-12]. In the kind of excitation transfer process under consideration, a de-excitation between the two states that occurs with phonon exchange can be coupled to an excitation at another site, provided that a phonon is exchanged with a common phonon mode. The excitation energy at one site in such a process is transferred to another site, and hence local energy and momentum conservation is violated at each.

There is nothing to prevent such a process from occurring in nature in principle. There is no reason to hold local energy and momentum conservation inviolate in atomic and molecular processes, since excitation transfer reactions can, and do, occur. To maintain local energy and momentum conservation to be an inviolate foundation in nuclear physics requires then a positive statement that there can not exist a viable excitation transfer mechanism. We are aware of no such arguments presented in the nuclear physics literature. If we wish to include excitation transfer reactions into the spectrum of possible nuclear reactions, then we need only revise the foundation so that global energy and momentum is always conserved, and that local energy and momentum conservation holds in all processes in which excitation transfer does not occur.

Anomalous energy exchange

Having transferred the reaction energy to another site does not solve the problem, since one might expect a rapid decay of the associated excitation energy into energetic products. We require a second mechanism which is capable of converting this large energy quantum into a very large number of smaller quanta.

As it turns out, one does not have to dig very deeply into the literature to find examples in which such effects are studied, although not to the degree that we require. In studies of the spin-boson problem there occur level anticrossings at which two levels mix, where one of the levels differs by one unit of two-level system energy and a large number of oscillator quanta. This is the most basic quantum mechanical model in which a two-level system with a large transition energy is coupled linearly to an oscillator that has a small characteristic energy, and one finds that energy can be coupled freely between them (albeit at a slow energy exchange rate) as soon as the coupling is increased to modest levels.

The mechanism through which this occurs in the spin-boson problem is straightforward. The interaction term in the spin-boson Hamiltonian couples one two-level system excitation or de-excitation to the creation or annihilation of an oscillator quantum. At modest coupling strength there occur frequent excitations and de-excitations of the two-level system, with oscillator quanta exchanged in association with each transition. In the event that more than one two-level system is involved, then excitation transfer reactions occur, and once again there is energy exchange with the oscillator associated with each excitation transfer reaction.

Spin-boson type model

We can implement these ideas into an idealized model in which two sets of two-level systems are coupled to a common oscillator. The associated Hamiltonian is [10-12]

$$\hat{H} = \Delta E_1 \frac{\hat{S}_z^{(1)}}{\hbar} + \Delta E_2 \frac{\hat{S}_z^{(2)}}{\hbar} + \hbar\omega_0\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right) + V_1\left(\hat{a}^\dagger + \hat{a}\right)\frac{2\hat{S}_x^{(1)}}{\hbar} + V_2\left(\hat{a}^\dagger + \hat{a}\right)\frac{2\hat{S}_x^{(2)}}{\hbar}$$

In this model, one set of identical two-level systems has a transition energy ΔE_1, and the other has a transition energy of ΔE_2. An oscillator is included with a characteristic energy $\hbar\omega_0$. Linear coupling between each set of two-level systems and the oscillator are included.

We have studied this model in the limit that the oscillator energy is low compared to the two-level system energies, and also in the limit that the oscillator is highly excited. The energy levels away from resonances can be parameterized according to

$$E_{n,M_1,M_2} = \Delta E_1(g_1)M_1 + \Delta E_2(g_2)M_2 + \hbar\omega_0\left(n + \frac{1}{2}\right) + \cdots$$

Since we are discussing a coupled quantum system in which the coupling may be strong, one might not expect that things work so simply. In essence, the coupled system seems to behave as if each set of two-level systems behaves almost as if they are uncoupled, but at an increased energy. The dimensionless coupling strengths in this case are

$$g_1 = \frac{V_1\sqrt{n}}{\Delta E_1} \qquad g_2 = \frac{V_2\sqrt{n}}{\Delta E_2}$$

The dressed energy $\Delta E(g)$ is given approximately by

$$\Delta E(g) = \frac{\Delta E}{\pi}\int_{-\sqrt{\varepsilon}}^{\sqrt{\varepsilon}}\sqrt{\frac{1 + 8g^2y^2/n}{\varepsilon - y^2}}$$

We have studied anomalous energy exchange within this model, in which energy from a two-level system is turned into oscillator energy and then back. If parameters of the model are selected such that a resonance is present for all the M_2 states such that

$$\Delta E_2\left(g_2\right) \;=\; \left(2k+1\right)\hbar\omega_0$$

then one can develop analytical solutions for the energy exchange. The resonance condition involves matching the dressed two-level transition energy $\Delta E_2(g_2)$ to an odd number of oscillator quanta due to selection rules. One finds solutions of the form

$$\left\langle M_2 \right\rangle \;=\; M_0 \cos^2\left(\Omega t\right)$$

where M_0 is a constant and where the frequency Ω is given approximately by

$$\Omega \;=\; \frac{\sqrt{2}\,g\hbar\omega_0}{\sqrt{n}}\,\frac{1}{\pi}\int_{-\sqrt{\varepsilon}}^{\sqrt{\varepsilon}} \frac{\cos\left\{ \dfrac{\Delta E}{\hbar\omega_0}\displaystyle\int_0^y \sqrt{\dfrac{1+8V_2^2 x^2/\Delta E_2^2}{\varepsilon - x^2}} \right\}}{1+8\left(\dfrac{V_2 y}{\Delta E_2}\right)^2}$$

where ε=2n+1. The goodness of this approximation can be seen in the results presented in Figure 1.
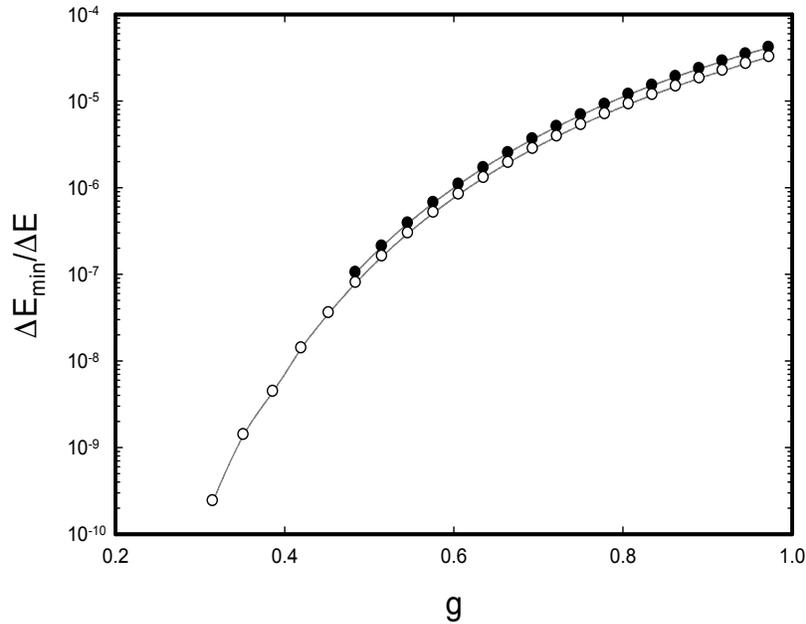
Figure 1 – Minimum energy splitting for anomalous energy exchange relative to the two-level transition energy in the case of $\Delta E_2 = 31\hbar\omega_0$. Solid circles, exact result from direct numerical solution; open circles, analytic estimate.

We have also found excitation transfer solutions within the model. It is even possible to find model parameters in which excitation transfer and energy exchange both occur [12]. Results for such a model are illustrated below in Figure 2. In this model, the system is initialized in a state in which the two-level system (in system 1) is excited, and none in system 2. Excitation transfer then occurs, producing a state in which the two-level of system 2 is excited, and none in system 1. In this model, this excitation transfer is accompanied by the exchange of 4 oscillator quanta. Next, energy exchange occurs so that the two-level system energy is converted to oscillator energy (in this case, 63 oscillator quanta per dressed two-level system energy of the second set). The overall process is coherent, and one sees that it reverses, and will repeat cyclically.
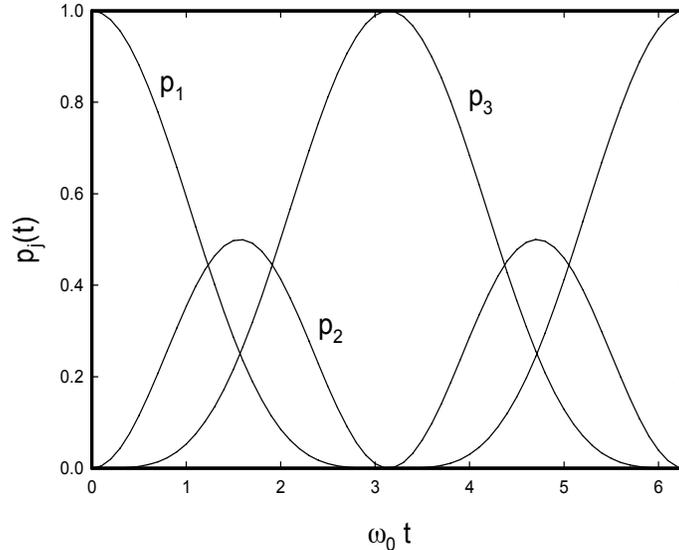
Figure 2 – Probabilities for excitation transfer followed by energy exchange under conditions where all states are degenerate in the absence of coupling, and when the coupling strengths are matched (from the example given in Ref. [12]). In the first state (with probability p_1), only the first two-level system is excited; in the second, only the second two-level system is excited; and in the third, neither two-level system is excited. The frequency ω_0 in this case is $\Delta E_{min} / 2\hbar$.

These models are interesting in that they exhibit all of the mechanisms required to support a model for excess heat production in the Fleischmann-Pons experiment. However, a study of these models shows that these effects can be seen only when precise resonances are present, and the associated rates are slow. In essence, this model is too weak. We can demonstrate excitation transfer, but the excitation transfer rate in this model is too slow to account for experiment. We can demonstrate anomalous energy exchange, but the rate at which energy is exchanged between the two systems is again too slow to account for experiment. We require a modification of the model in which the excitation transfer rate is greatly accelerated, and in which the anomalous energy exchange rate is also greatly accelerated. We also require a modification in which the requirement for precise resonances is relaxed.

Augmentation with loss processes

In the event that the two-level system energy is in the MeV range, and the oscillator corresponds to phonon modes in the lattice, one would expect that new loss channels should be present associated with the oscillator. For example, coupling between the two-level systems and oscillator means that the oscillator has available large energy quanta through the off-resonant coupling inherent in the model. Hence, if the lattice has access to any loss channels at the

energy of the two-level systems, we would expect that decay through these channels should be allowed. Such processes include indirect nuclear disintegration and energetic electron ejection.

The existence of such loss channels has a big impact on the models under discussion as we shall see. Hence, we need to augment the models in order to include them. To do so, we augment the idealized spin-boson model to include such loss channels. We write [10-12]

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

To include loss effects directly into a Hamiltonian involves technical issues, since the Hamiltonian itself must be conservative. Our approach is to divide the associated Hilbert space into sectors, and then focus on the sector of interest. Loss terms mediate transitions out of the sector of interest. In this way, the model Hamiltonian for the overall system is Hermitian, but the restricted Hamiltonian associated with a sector can include loss terms that are anti-Hermitian. Loss of probability from the sector limits the applicability of such a model, but is very useful for our discussion here.

Excitation transfer with loss

It is possible to use perturbation theory to estimate the excitation transfer rate in the limit of weak coupling. One way to do it is to select 6 basis states according to [12]

$$\phi_1 = |n\rangle|s,1/2\rangle|s,-1/2\rangle \qquad \phi_2 = |n-1\rangle|s,-1/2\rangle|s,-1/2\rangle$$
$$\phi_3 = |n+1\rangle|s,-1/2\rangle|s,-1/2\rangle \qquad \phi_4 = |n-1\rangle|s,1/2\rangle|s,1/2\rangle$$
$$\phi_5 = |n+1\rangle|s,1/2\rangle|s,1/2\rangle \qquad \phi_2 = |n\rangle|s,-1/2\rangle|s,1/2\rangle$$

We have an initial basis state ϕ_1 with one two-level system excited in the first set, and none in the second. We have a final basis state ϕ_6 in which one two-level system is excited in the second set, and none in the first. The remaining states are intermediate states in a transition from the initial to the final. The expansion coefficients c_j satisfy

$$Ec_1 = H_1 c_1 + V_1\sqrt{n}c_2 + V_1\sqrt{n+1}c_3 + V_2\sqrt{n}c_4 + V_2 c_5$$
$$Ec_2 = H_2 c_2 - \frac{i\hbar\Gamma}{2}c_2 + V_1\sqrt{n}c_1 + V_2\sqrt{n}c_6$$
$$Ec_3 = H_3 c_3 - \frac{i\hbar\Gamma}{2}c_3 + V_1\sqrt{n+1}c_1 + V_2\sqrt{n+1}c_6$$
$$Ec_4 = H_4 c_4 + V_2\sqrt{n}c_1 + V_1\sqrt{n}c_6$$
$$Ec_5 = H_5 c_5 + V_2\sqrt{n+1}c_1 + V_1\sqrt{n+1}c_6$$
$$Ec_6 = H_6 c_6 + V_2\sqrt{n}c_2 + V_2\sqrt{n+1}c_3 + V_1\sqrt{n}c_4 + V_1\sqrt{n+1}c_5$$

Note that loss occurs here only for basis states 2 and 3, which have basis energies lower than the initial state, and hence which are potential final states for an energetic decay process. Loss in this simple model is included as a constant.

We can eliminate the expansion coefficients of the intermediate states to obtain coupled equations of the form

$$Ec_1 = H_1 c_1 + \Sigma_1(E) c_1 + V_{16}(E) c_6$$
$$Ec_6 = H_6 c_6 + \Sigma_6(E) c_6 + V_{61}(E) c_1$$

The indirect coupling coefficient $V_{16}(E)$ is

$$V_{16}(E) = V_1 V_2 \left[\frac{n}{E - H_2 + i\hbar\Gamma/2} + \frac{n+1}{E - H_3 + i\hbar\Gamma/2} + \frac{n}{E - H_4} + \frac{n+1}{E - H_5} \right]$$

If there is no loss, then this indirect coupling coefficient becomes

$$V_{16}(E) \rightarrow \frac{2\hbar\omega_0 V_1 V_2}{\left[\Delta E_1(g)\right]^2 - (\hbar\omega_0)^2}$$

If the loss is taken to be infinite, then this indirect coupling coefficient becomes

$$V_{16}(E) \rightarrow -\frac{2 V_1 V_2 \Delta E_1(g_1) n}{\left[\Delta E_1(g_1)\right]^2 - (\hbar\omega_0)^2}$$

The loss is seen to have a dramatic impact on the indirect coupling between the initial and final states. In the absence of loss, the coupling is weak due to destructive interference effects. The inclusion of loss breaks this destructive interference, yielding a much increased coupling.

Connection with experiment

The inclusion of loss in these models increases the excitation transfer rate as well as the rate for anomalous energy exchange. The rates that result are very large, and sufficiently large that they can support models for excess heat in the Fleischmann-Pons experiment [10,11].

Let us examine briefly the rate associated with excitation transfer in the strong coupling regime (in which excitation transfer is the bottleneck). In this case, the maximum rate at which excitation transfer can proceed in the model on a per deuteron pair basis is

$$\gamma \rightarrow \frac{2 V_1 \sqrt{n}}{\hbar}$$

It is possible to develop an estimate for this rate in terms of the coupling matrix element associated with a direct transition from local molecular D_2 states into ^{4}He states, mediated by phonon exchange. In this case, transitions are driven by compressional modes, and the largest matrix elements are expected for S=2 (nuclear spin) molecular states. The associated rate can be approximated crudely by

$$\gamma \rightarrow \frac{v}{\hbar} e^{-G} \sqrt{\frac{vol_{nuclear}}{vol_{molecular}}}$$

Where e^{-G} is the Gamow factor associated with tunneling, where the square root includes the ratio of volume factors for a transition from a molecular state to a nuclear state, and where v is the Gamow factor and volume factor reduced matrix element that we anticipate to be O(MeV). The reaction power associated with a reaction rate of this magnitude is

$$P \rightarrow N[D_2]\Delta E_1 \sqrt{\frac{vol_{nuclear}}{vol_{molecular}}} e^{-G}\frac{v}{\hbar}$$
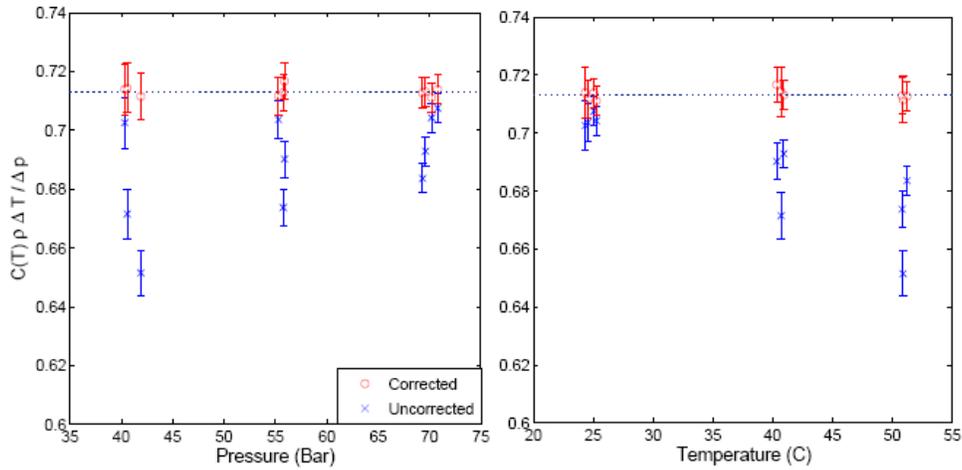
Figure 3 – Measure of the thermal gain across the cell (thermal output over pdV work). Blue data points – uncorrected data as taken; red points – corrected data. More than 28% of the mechanical power is lost going through the cell to loss mechanisms including phase changes and other effects. Plots from Figure 10-7 of [13].

Inserting estimates for Gamow factors and volume factors associated with molecular D_2 in vacuum leads to the conclusion that the associated power is quite small (attowatts) for a plausible choice of the number of D_2 molecules involved in a Fleischmann-Pons experiment. However, low-energy deuteron beam experiments on deuterated targets shows that screening effects dramatically increases the tunneling factors in metals in general and especially in Pd in particular. Excess power estimates at the watt level can be obtained with screening factors much more modest than those consistent with experiment. We conclude from this that the generalized spin-boson models augmented with loss are capable of describing excess heat in the Fleischmann-Pons experiment, but that accurate predictions of excess heat will require accurate screening models for D_2 in the vicinity of vacancies. Nevertheless, these models illuminate much about the mechanisms and requirements for excess heat production.

Connection with the Yang-Koldomasov experiment

We discussed the Yang-Koldomasov experiment in the last RLE report. Observations of this experiment made in early 2005 during demonstrations done in Canada indicated that more thermal energy was observed as output than electrical energy as input by a factor of perhaps 5-7. We hosted a confirmation test of a Yang-Koldomasov experiment last year in our lab at MIT in the hope of seeing excess heat production and other anomalies attributed to the machine.

With considerable help from FRC, we ran the machine at MIT, and studied thermal measurements in order to develop the capability to perform accurate calorimetry. From these studies, we found that it was possible to achieve accuracy at the few per cent level, based on several independent measurements. No excess heat was observed during these tests (see Figure 3 for an example of data taken). In the demonstrations done previously in Canada, very strong transverse arcs were observed which were recognized as being consistent with ion discharges following fast electrons into the plastic cell surrounding the nozzle. These were absent in the tests done at MIT. Work continues at FRC to explore different cell configurations and operating regimes.

We described models in last year's report that appeared to be relevant to the Yang-Koldomasov experiment. These models are very similar to those under discussion here, except that we had been using weak coupling estimates for the reaction rates, rather than estimates for the strong coupling regime found subsequently and discussed here. According to the earlier models, the weak coupling estimates gave fast reaction rates with nonlinear dependencies on constituent concentrations, with the reaction rates estimates based on unscreened Gamow factors computed to be very large. In the strong coupling limit, the model works differently qualitatively, and leads to quite small reaction rates for the parameter regime under discussion. Because the strong coupling rates are so much less, the screening effects due to metals appear to be a prerequisite for excess power production, which will carry over to p+d reactions as well. Consequently, according to the new model, there must be a suitable metal host for the reactants either in the oil, or in the metal surfaces in contact with the flow.

Laser beating experiments

Some years ago, Letts and Cravens demonstrated that a weak diode laser operating near 2 eV could stimulate excess heat bursts in Fleischmann-Pons cells that they were studying [14]. In these experiments, the excess power increment was one to two orders of magnitude greater than the incident laser power. Subsequent work showed that the effect only occurred when the laser polarization was TM, which could couple to compressional surface plasmon modes.

No. 662a1 Excess Pwr & Beats Vs Time
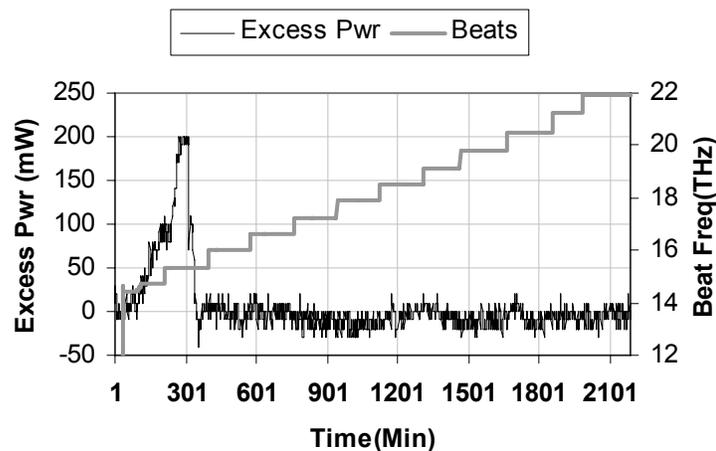
Figure 4 – Excess power and beat frequency as a function of time, taken from Figure 7 of Ref. [15]. An excess heat event was stimulated at a laser beat frequency near 14 THz.

More recently, Letts and Cravens have pioneered new experiments in which excess heat appears to respond to the frequency of the laser beating at three frequencies in the optical phonon region between 8-20 THz [15]. These new experiments are the first to implicate optical phonon modes directly with excess heat in the Fleischmann-Pons experiment. This helps to provide a direct connection between the models under discussion and experiment. We note that the rate of anomalous energy exchange is much higher in the case of a higher energy oscillator, so that this beating effect may shed light most importantly on this part of the overall process.

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