

Research Article

Theory and Experiments in Condensed Matter Nuclear Science

Peter L. Hagelstein*

Massachusetts Institute of Technology, Cambridge, MA, USA

Abstract

Excess heat in the Fleischmann-Pons experiment implies the possibility of small-scale source of clean nuclear energy, with the potential to address climate change, finite oil supply, and clean water problems. Following the announcement on March 23, 1989, it took on the order of 40 days for the effect to be rejected, which has led to a bifurcation in which researchers in the field has studied the effect, and mainstream science general does not accept the existence of it. We consider the theoretical picture long studied in our group, and connections with experiment. The model involves many steps, and we have made an effort to connect as many of the theoretical steps as possible to experiments that seem to be closely related.

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1. Introduction

At the LENR Workshop in honor of my friend Srini, I submitted a video presentation that focused on a variety of experiments in connection with theoretical issues and mechanisms [1]. This presentation might form the basis of a review article at some point in the future, which might discuss a variety of experiments along with proposed interpretations that might be helpful in providing a consistent picture and help point out connections between them. The development of such a review at the moment is hindered due to the realities of the ongoing pandemic (I do not have approval to go to my office where my books and papers are). Instead it may be that a more workable project involving a brief discussion of thoughts on experiment and theory might be useful to the community.

2. Significance of the Fleischmann-Pons experiment

Fleischmann and Pons described an electrochemical experiment in their March 23, 1989 announcement in which large amounts of thermal energy was observed in the absence of commensurate chemical products. It was conjectured that the source of the energy produced was nuclear, in spite of the absence of commensurate energetic nuclear products [2], [3]. If real, this significance of this claim cannot be overstated. A correlation between the energy produced and ^4He detected as a product is discussed below, but this helium appears to be born with very little energy.

*E-mail: plh@mit.edu

In conventional nuclear fission and nuclear fusion processes, when significant nuclear energy is released, it is expressed in terms of energetic nuclear particles. In the case of nuclear fission there are observed energetic MeV fission products, and for nuclear fusion there are observed energetic MeV fusion products. In the Fleischmann-Pons experiment as is presently understood, the claim is that nuclear energy is produced without commensurate energetic MeV products. From a scientific perspective this implies a fundamentally different process is involved, in contrast to conventional (quantum mechanically incoherent) fission and fusion.

From a societal perspective this experiment suggests the possibility of a new small scale clean nuclear energy source. Although still not conclusively proven, many think that the source of the energy in PdD experiments is the mass difference energy between D_2 and 4He , which would suggest a potentially inexhaustible and inexpensive fuel source. New energy technology taking advantage of this effect could potentially provide solutions to some of the major problems humanity currently faces, including energy supply, clean water, and global warming. The extremely high energy density associated with a small scale clean nuclear energy source would have the potential to impact home heating and cooling, industrial energy, transportation, robotics, and space travel.

3. Fleischmann-Pons in the media after the announcement

Substantial public interest in the Fleischmann-Pons experiment followed the initial announcement, with daily coverage in the media. This was viewed by some as problematic at the time. In the US much scientific research is supported by government funding, which places a premium on maintaining public support for science. Because of this there has been an interest by some scientists in maintaining an appropriate public image of scientists and of the scientific endeavor, resulting in a steady diet of largely positive news reports, documentaries, and science-related programs in the media. There was concern about the increasing public focus on the Fleischmann-Pons experiment, which many physicists thought involved a claim inconsistent with the laws of physics.

One possible scenario might have been to take the time to study and to understand the experiment, which with substantial support from the government funding agencies might have been done over the course of perhaps five years. If at the end of some years of investigation the result was negative, science itself would likely not have been perceived as positively by the public as a result of the associated disappointment.

The expectation among some senior physicists was that it would be found that the basic claim was a result of errors made in carrying out the experiment, largely since the dramatic claim was at odds with previous experience with nuclear energy generation. This suggested a “fail-fast” strategy for cold fusion which would remove it from the center of public attention sooner rather than later.

4. The Baltimore APS meeting

At the Baltimore APS meeting roughly 40 days after the initial announcement, skeptical scientists had their day, and also carried the day. The excess heat experiment could not be reproduced [4]. A theoretical model was presented which suggested that, because of the Coulomb barrier, fusion would not be expected to occur at a rate needed to be relevant to experiment [5]. The impact of this meeting on scientific discourse cannot be overstated. Prior to this meeting, there were frequent discussions about the experiment, the claimed results, the significance, and on how it might work. After this meeting, this stopped (except when cold fusion enthusiasts gathered). The Fleischmann-Pons experiment had effectively been knocked out of science.

In light of this pivotal event, there are some questions which might be asked.

As a technical description of the Fleischmann-Pons experiment and the associated experimental protocol used by Fleischmann and Pons had not been made public, exactly what experiments were being done that carried such weight? Could it really be argued that an experiment was not reproducible, prior to anyone other than those immediately associated with Fleischmann and Pons having relevant technical details about the experiment?

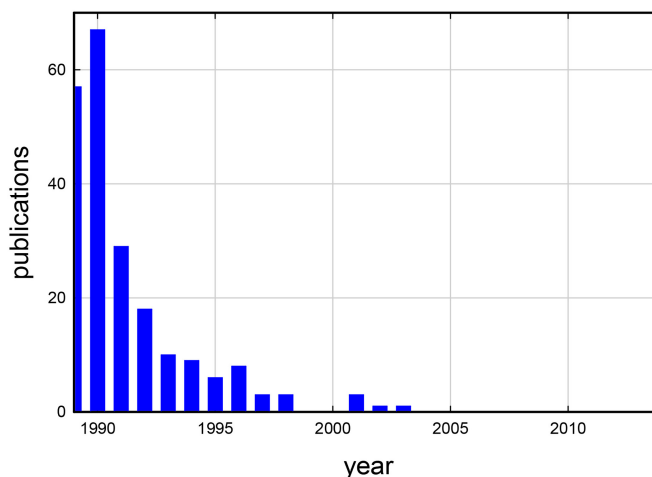


Figure 1. Histogram of the 217 experimental papers with negative results for excess heat in the Britz bibliography.

Subsequently it was understood that the Fleischmann and Pons experiment of that era typically lasted on the order of two months, so that there was not sufficient time between the announcement and the Baltimore APS meeting for a single experiment of theirs to be completed.

There was a focus on a model for the fusion rate for molecular D_2 , which would result in $p+t$ and $n+{}^3\text{He}$ as primary fusion products. Due to the difficulty in tunneling through the Coulomb barrier, the associated fusion rate per molecule was on the order of $10^{-64} \text{ sec}^{-1}$. This fusion rate was sufficiently slow that there emerged confidence that the many orders of magnitude required to account for the Fleischmann-Pons experiment could not be made up in any scenario.

In retrospect it might be pointed out that the claim made by Fleischmann-Pons was for excess heat of nuclear origin without commensurate neutron production. As the mechanism under discussion at the Baltimore APS meeting would have produced neutrons had it occurred at substantial rates, it would seem that ruling it out could be argued to be in agreement with the Fleischmann-Pons experiment. At this point the experimental evidence suggests that neutron emission is not correlated with excess heat in the Fleischmann-Pons experiment, with an upper limit near 1 neutron per 100 Joules of energy generated. There is no reason to believe that the incoherent fusion decay of molecular D_2 occurs at any significant rate in a Fleischmann-Pons experiment operating in a regime where substantial excess heat is produced.

A better theoretical argument might have involved rate estimates for a theory that actually had something to do with the experimental claim. If no commensurate neutron production was observed in connection with the thermal energy measured, then presumably it would be appropriate to make arguments involving a model that did not involve neutron production. In light of subsequent work, arguments made based on any model that predicted commensurate energetic MeV nuclear particles would not have been relevant.

5. Preponderance of the evidence

There were subsequently hundreds of publications of experiments in which negative results were reported. In 2014 I downloaded the extensive bibliography that had been posted by Dieter Britz [6], which included references for many journal publications of experimental results that were negative. A histogram of publications by year with negative experimental results for excess heat is shown in Figure 1. With so many experiments having been done, it could be

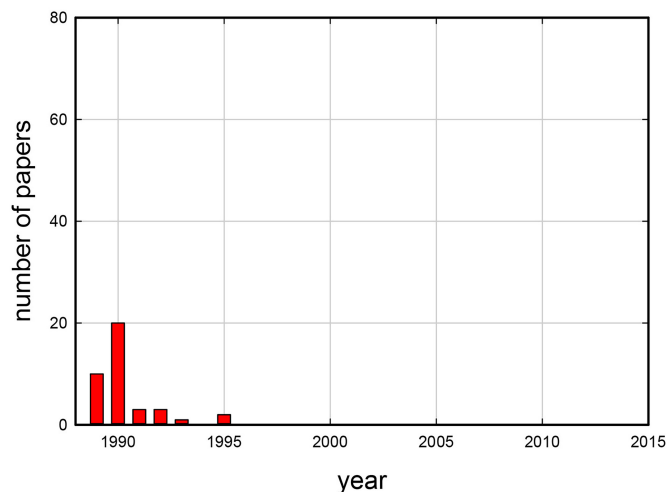


Figure 2. Histogram of the 39 experimental papers with negative results for excess heat in the Britz bibliography, where an electrochemical experiment involving Pd was done and a temperature measured.

argued that independent of whatever shortcomings there may have been with the early dismissal of the Fleischmann-Pons experiment at the Baltimore APS meeting, the experiment had gotten looked at and found wanting.

I recall discussions with some physicists at MIT in the early 1990s in which the Fleischmann-Pons experiment was discussed. It was explained to me that there were hundreds of experiments done at reputable labs, and that nobody was seeing anything. Moreover, that if I wanted to convince anyone that there was anything to the Fleischmann-Pons experiment, that what would be required would be for every one of these negative experiments to be looked at to identify what was done wrong.

One approach to this might be to work with the body of papers included by Britz. As the issue under discussion is whether the Fleischmann-Pons experiment produces excess heat, as a restriction we might include only papers in which an electrochemical experiment involving Pd was done and a temperature measured. A histogram of this subset (including 39 papers) is shown in Figure 2.

In the early meetings on excess heat in the Fleischmann-Pons experiment there was discussion of the importance of achieving high D/Pd loading. An oblique reference to this is apparent in retrospect in their first paper [2] in the second paragraph, where a high value 0.8 eV for the chemical potential of D in Pd. The connection between D/Pd loading in the SRI experiments and in other experiments is discussed briefly below, and is often mentioned in the early papers in our field.

This provides motivation to examine the early negative results to see whether the researchers were aware that a high D/Pd loading was important. An initial restriction to include papers where D/Pd loading is mentioned reduces the total to 22 papers. In only 7 of these papers describing negative experimental results was the D/Pd loading reported to have reached 0.83.

At SRI two different loading requirements were found – that if a cathode did not reach a D/Pd loading near 0.95 or higher while being charged, then it did not show excess heat; and that when excess heat was observed, a threshold for excess heat production in the vicinity of a loading of 0.83 was seen. In only 3 of the negative experimental papers was a loading as high as 0.90 reported.

From our perspective, the best of these papers was the publication of Green and Quickenden (1995) [7], in which a maximum D/Pd loading of 0.96 (as determined from the same resistance ratio measurements used at SRI) was reported, and no excess heat was seen with an experimental error at the level of $\pm 1.5\%$. We note that in the SRI experiments it was found that current ramps were often effective at stimulating an excess heat burst (and that excess heat appeared infrequently under constant current conditions) [9]. In the Green and Quickenden experiments, a protocol was used in which the current was switched suddenly from low to high current and held constant at high loading – no current ramps were employed.

In subsequent years there was an expansion of the types of cathodes studied. A drawback of a “large” rod cathode is that it takes a long time to load at low current density, where a thin foil cathode can be loaded much more quickly. Very high loading can be achieved in these cathodes. The Energetics/SRI/ENEA/NRL collaboration saw excess heat in many such thin foil cathodes, in some cases exploiting the “superwave” current protocol [10]. However, no excess heat was seen in experiments at Coalescence where similar thin Pd foil cathodes were used.

6. Is it real?

It is astonishing that in 2021 the question of whether excess heat in the Fleischmann-Pons experiment is real or not remains an issue.

In the same Britz bibliography mentioned above, one can find 49 positive excess heat results in Fleischmann-Pons experiments which could reasonably be compared to the 39 negative results restricted as discussed above. Yet this appears to have had no impact on mainstream science. By now there have been hundreds of positive excess heat results reported within our community. While there is disagreement on nearly every important point in our field (such as how it works), one thing that most everyone agrees on is that the excess heat effect is real (and worthy of study). This view is not shared in mainstream science. The large body of research carried out, reported on at conferences, written up and in many cases published, simply appears to carry no weight.

There have been many discussions in our field at conferences and in private, as to what would be needed to change the situation. One line of argument has it that what is needed is a “lab rat” experiment that anyone can do, with kits to be developed and shared with mainstream labs. Another line of argument is that things will change when a commercial product becomes available. In the past I have opined that if we managed to figure out how it worked in detail, then things would change.

In retrospect I am wondering whether the problem lies elsewhere. For example, the initial onslaught involved scientific arguments shaky at best, and over-reaching. It was argued confidently, forcefully, and repeatedly that the excess heat effect was not reproducible. This was at a time when Fleischmann and Pons were reproducing their own results. And later on many others observed excess heat. In fact the excess heat effect can be observed, now, as was the case in 1989. Yet this very loud and confident opinion has saturated mainstream science for decades, and there has been no practical way to respond so as to have a significant impact.

The problem in my view is a political and sociological one. A question of science can presumably be answered making use of science. But making use of science in connection with excess heat in Fleischmann-Pons experiments has demonstrably had no effect for three decades and more, largely because the issue was never a scientific one.

The excess heat effect is real. Mainstream science is convinced otherwise, and officially has no interest in the problem. And this has been the case for more than three decades, and could easily remain the status quo for a great many more decades.

7. How does it work?

One should be interested in the question of how it works. An experiment which produces large amounts of thermal energy from clean nuclear reactions suggests the possibility of new commercial technologies that would revolutionize

modern society, along with solving quite a few of the biggest problems facing humanity. This would seem to provide motivation for wanting to understand what is going on under the hood.

Although there have been hundreds of theory papers in our field, there is no agreement as to what is going on microscopically.

It seemed clear (to me) back in 1989 that the only kind of theoretical approach that could have anything to do with the experimental claim was if there were a way for fusion to occur as a coherent process. In quantum mechanics, the decay of one state to a continuum leads to a model studied first by Dirac [8] (the Golden Rule) which in most cases leads to (incoherent) exponential decay. The dynamics of a single state coupled to another single state at the same energy leads to coherent dynamics (Rabi oscillations). An issue is that coherent dynamics can be much faster than incoherent dynamics, especially if the coupling matrix element is weak (as is the case when tunneling through a Coulomb barrier is involved).

A coherent model is needed which starts with a molecular D_2 species somewhere in the lattice, and ends with a ^4He nucleus at the same place, with the energy split up into little pieces and deposited elsewhere. Over the years a very large number of models fitting this description have been studied, leading to an evolution to ever more complicated versions.

In the most recent versions of the scheme, the first step involves the transfer of the large $D_2/{}^4\text{He}$ (24 MeV) quantum to a reasonably stable highly excited state in a nucleus in the lattice (either host lattice or impurity). This first step would undergo collective enhancement if there were a lot of $D_2/{}^4\text{He}$ transitions coupling to a common highly excited THz vibrational mode, with the excitation coupled to many identical excitations in the host lattice all interacting with the same highly excited mode. A second step might involve a subdivision, in which the excitation was split in half and transferred to reasonably stable states of nuclei in the lattice at roughly half the energy of the $D_2/{}^4\text{He}$ transition. Once again there are collective enhancements if all of the excitation of many $D_2/{}^4\text{He}$ transitions proceed through a small number of common reasonably stable states of identical nuclei in the host lattice. If this subdivision continues the original large quantum is ultimately split among a large number of nuclei in low-lying reasonably stable states in the keV or 10s of keV range. In the models under discussion, the energy gets transferred to vibrations through massive multi-quantum down-conversion.

A reviewer has asked about how the $D_2/{}^4\text{He}$ transition works. In the simplest picture, molecular D_2 can be viewed as an excited state of ${}^4\text{He}$. In this case single phonon exchange can mediate a transition, as long as the selection rules are satisfied. Photon-mediated exchange is known in the case of accelerator experiments in which two deuterons collide leading to gamma emission [11], and back in 1989 this was recognized as implying that radiative decay of molecular D_2 to ${}^4\text{He}$ is expected. Phonon exchange is conceptually similar [12], except that phonon modes are restricted to vibrational energies below about 400 meV, so that a single-phonon decay process is not possible. Instead one would expect higher-order processes to occur, such as excitation transfer [13].

8. Excitation transfer of the 24 MeV quantum

In the model outlined above, excitation transfer of the large 24 MeV quantum from the $D_2/{}^4\text{He}$ transition to produce a highly excited and reasonably stable state in a Pd nucleus is proposed as the first step in the excess heat process. When things work properly this excitation is quickly transferred elsewhere via subdivision, rendering it essentially unobservable. However, in experiments below threshold for excess heat production, the subsequent excitation transfer is not fast, allowing for the possibility of seeing the decay products.

Some of our earliest speculations about mechanism focused on an experimental observation by Chambers et al in which ion bombardment of Pd with deuterons led to a few counts (possibly alphas) near 20 MeV [14]. This would be consistent with an excitation transfer process that we might write as



Table 1. Alpha and neutron energies resulting from the stable Pd isotopes.

isotope	E_α (MeV)	E_n (MeV)
^{102}Pd	20.86	13.14
^{104}Pd	20.43	13.73
^{105}Pd	20.16	16.59
^{106}Pd	19.84	14.15
^{108}Pd	19.25	14.49
^{110}Pd	18.71	14.91

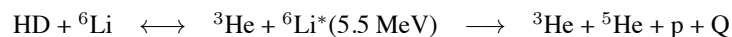
with the alpha energy Q listed in Table 1. In subsequent experiments the group was not able to replicate the effect, casting doubt on the initial observations. In our lab we are working with a deuteron beam seeking this reaction among others.

The highly excited state in Pd would presumably have proton and neutron decay channels, leading to energetic neutrons. Low-level energetic neutron emission has been reported in the co-dep experiments of Mosier-Boss and coworkers [15], which may be consistent with this reaction. Energetic protons and alphas above 10 MeV were described by Lipson and coworkers [16], which also may be consistent with this kind of process. A discussion of phonon-mediated nuclear excitation transfer and incoherent decays is given in [13].

Phonon-mediated nuclear excitation transfer is fundamental to this first step in the model, as well as to subsequent steps. Consequently it is important in connection with the discussion to understand whether it occurs in nature. In our lab we are working on basic excitation transfer experiments involving excited nuclear states of ^{57}Fe produced by the decay of ^{57}Co [17]–[19], where preliminary results suggest that under non-optimized conditions individual transfers can occur on a picosecond time scale.

9. Excitation transfer of the 5.5 MeV quantum

The $\text{HD}/^3\text{He}$ transition energy is much lower (5.5 MeV), with the potential to produce alpha emission only in a few low- A nuclei. We have drawn attention to



as a candidate for the proton signal reported by Lipinski and Lipinski [20].

We consider briefly the situation for excitation transfer of the 5.5 MeV quantum to nearby levels in the nickel isotopes in a NiH excess heat experiment. Known energy levels for the nickel isotopes are available in the NUDAT2 database which can be compared with the 5.493478 MeV mass difference energy, as shown in Figure 3. We see that the density of states is highest for ^{61}Ni , and that there are states in the general vicinity of the $\text{HD}/^3\text{He}$ transition energy. A close-up plot of the states between 5400–5600 keV are shown in Figure 4, which shows that the closest levels are within about 5 keV, which seems not to be particularly close.

Whether any of these levels might be candidates for the first transition depends on whether single phonon exchange can occur with energy transfer substantially different than a phonon quantum. If excess heat is being produced and if the down-conversion models are relevant, then the transfer of a few keV seems possible. If the system is starting from scratch, then there would be no extra energy available in the case of single phonon exchange, in which case none of the nearby levels would be candidates. We would need levels (presumably in impurity nuclei) much more nearly resonant.

This motivates a search of the database to seek levels more nearly resonant. Results are given in Table 2. The closest level listed in the database is ^{116}Sn at 5493.2 keV, which looks to be within a few hundred eV of the $\text{HD}/^3\text{He}$ transition energy.

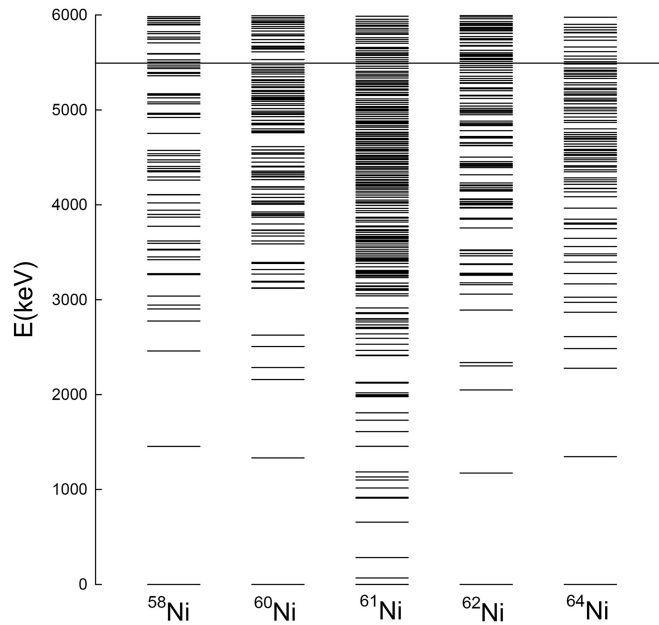


Figure 3. Energy levels of the stable Ni isotopes compared with the HD/³He transition energy.

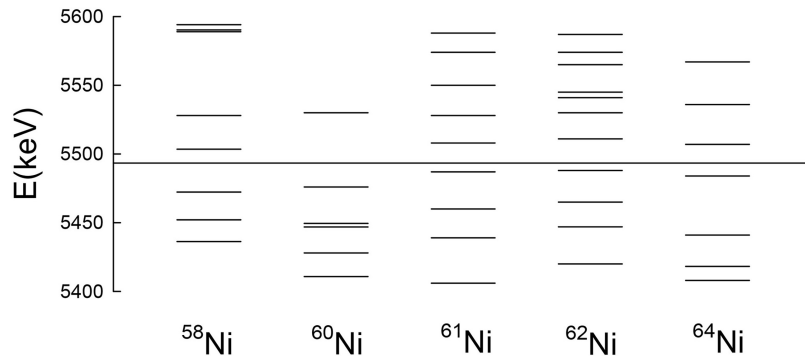


Figure 4. Close up of energy levels of the stable Ni isotopes compared with the HD/³He transition energy.

Unexpectedly, we see that ¹³⁷Cs has an excited state that is on the order of 700 eV above the HD/³He transition energy. The immediate thought is in connection with the Piantelli experiment, where a modest gamma signal at 661 keV was seen correlated with excess heat production in their gas loaded NiH experiment [21]. Perhaps there was a small amount of ¹³⁷Cs contamination in the Ni, so that when excess heat is produced much of the energy mismatch could be made up, resulting in near-resonant mixing that results in a much accelerated beta decay resulting in a gamma at 661.7 keV.

Table 2. Isotopes with excited states near 5493.5 keV.

AZ	$E(\text{keV})$
^{208}Pb	5490
^{37}Cl	5490.68
^{42}Ca	5490.77
^{50}V	5491
^{87}Rb	5491
^{74}Se	5491.2
^{206}Pb	5493
^{74}Ge	5493.1
^{116}Sn	5493.2
^{140}Ce	5494
^{137}Cs	5494.2
^{64}Zn	5495
^{186}Os	5495.5
^{91}Zr	5495.6
^{116}Sn	5495.91
^{51}V	5496
^{91}Zr	5496
^{142}Nd	5496

One wonders about a potential connection with the accelerated decay of ^{137}Cs in the experiments of Vysotskii, Kornilova and coworkers [22]. The associated picture would have the HD/ ^3He transition indirectly coupling with the 5494.2 keV state in ^{137}Cs to produce a modest (at most $35\times$) increase in the beta decay rate, where in the Piantelli experiment the energy mismatch could be made up leading to a more nearly resonant coupling with a decay rate larger by orders of magnitude.

10. First subdivision

From a theoretical perspective the down-conversion of the large 24 MeV quantum through subdivision seems compelling. However, we would like support from experiment to gain confidence in the theoretical picture. The first step in the proposal for excess heat is excitation transfer from the $\text{D}_2/{}^4\text{He}$ transition to a highly-excited reasonably stable excited state near 24 MeV in a host lattice nuclei (or impurity nucleus), and the connection to experiment was discussed above. The second step would involve a subdivision in which excitation transfer processes leads to two highly-excited nuclei with roughly 12 MeV excitation. We might expect a minor advantage for the associated coherent rate under conditions where the energy is split exactly in half; but it may probably be asking too much to have a reasonably stable state in just the right place. So more likely there are a few reasonably stable states near the half-energy that split the total.

The neutron energies that would result from a subdivision into two excited nuclei with exactly the same energy is shown in Table 3. It might be hard to distinguish neutrons with 2.34 MeV and 2.67 MeV energy from the 2.45 MeV dd-fusion neutrons in an experiment. However, neutrons near 4.78 MeV would be sufficiently separated as to be resolved as different. If we turn our attention back to an old conference proceedings paper by Takahashi and coworkers [23], we see excess neutron counts between 4 and 6 MeV, as well as counts below 2.45 MeV.

Table 3. Neutron energies for equal subdivision near 12 MeV.

isotope	E_n (MeV)
^{102}Pd	1.34
^{104}Pd	1.92
^{105}Pd	4.78
^{106}Pd	2.34
^{108}Pd	2.67
^{110}Pd	3.10

11. Further subdivision and the Gozzi experiment

Further subdivision in the scheme is proposed to down-convert the roughly 12 MeV quanta further down to the keV or 10s of keV level. The relative lack of metastable low-energy states in the Pd isotopes suggests that impurity nuclei may be important to get to low-lying nuclear excited states. One interpretation of collimated gamma emission in the Gozzi experiment [24] as a result of many subdivision processes, in which some of the large 24 MeV quantum ends up in metastable states in impurity silver isotopes. These include the 88.034 keV metastable state in ^{109}Ag , and the 93.125 keV metastable state in ^{107}Ag , as candidates. Collimated emission in this picture is a consequence of resonant excitation transfer among the different Ag nuclei in grains.

12. Down-conversion into vibrations

Once subdivision has reduced the large 24 MeV quantum into much smaller quanta associated with nuclear excited states in the keV and 10 keV range, the proposal is that (massive) down-conversion into THz phonons occurs. We have discussed previously models in which loss disrupts the destructive interference associated with down-conversion in spin-boson type of models [25], as well as the impact that off-resonant nuclear state energy shifts would have on the process [26]. The up-conversion and down-conversion effects under consideration would involve a great many individual non-resonant excitation transfer processes while maintaining coherence.

There are a variety of experiments in which collimated x-ray emission has been reported [27]-[30]. We have proposed the possibility of the inverse process up-conversion of THz phonons [31] as an explanation for such effects. This is also possible in the case of collimated gamma emission in the Gozzi experiment mentioned above. We have also proposed that massive up-conversion in fracture experiments may be responsible for elemental anomalies that have been reported (see [32] and references therein).

13. Phonon gain

According to the models the strongest effects should involve THz vibrations. Given the relatively small coherence volume for THz phonon modes (a few hundred Angstroms)³, this would limit the number of D_2/He transitions that interact, and the number of host metal nuclei that could interact. A much larger collective enhancement is possible in the event that there is phonon gain, which is consistent with the models. We have remarked previously that the Letts 2-laser experiment [33] provides indirect support for the notion of phonon gain (excess heat persists after the two lasers are turned off, where in 1-laser experiments the excess heat turns off when the laser turns off, consistent with a picture in which some of the nuclear energy goes into the vibrational modes to sustain them). More direct support for the proposal comes from the Raman experiments in which strong anti-Stokes lines are claimed [34].

14. Helium

According to Miles, Fleischmann was the first to observe helium in connection with cold fusion [35]. A nice review of the correlation of excess heat and helium production is given in [36]. In the model under discussion the source for the energy is the mass difference between D_2 and ${}^4\text{He}$, which suggests that there should be a relation between the energy produced and the number of ${}^4\text{He}$ atoms created

$$E_{xs} = N[{}^4\text{He}] \Delta M c^2$$

where the mass difference $\Delta M c^2$ is near 24 MeV. There are numerous experiments in which efforts to measure both energy and ${}^4\text{He}$ in the same experiment to test this. A problem associated with this kind of measurement is that when made, the helium becomes trapped in the metal, which in many experiments results in an under-estimate of the amount of ${}^4\text{He}$ produced.

There have so far been two measurements in which an effort was made to scrub out the ${}^4\text{He}$. One was in the M4 experiment run at SRI in the 1990s [37], and the other was done at ENEA Frascati (see the Laser-3 point in Figure 22 in [39]). In both cases the results were consistent with a 24 MeV mass energy to within experimental error. However, given the importance of this issue, more study of this relation is needed.

There has at this point been no equivalent result for the correlation between ${}^3\text{He}$ and excess heat in the NiH system involving an appropriate quadrupole mass spectrometer.

15. ${}^4\text{He}$ energy at birth

In an accelerator experiment when nuclei fuse, or more generally interact in some way, one can learn much about the reaction based on measurements of the reaction products. Energy and momentum conservation place strong constraints on the final state products. For example, in the case of the dd-fusion reaction for molecular D_2 at rest, energy and momentum conservation would require the 3.27 MeV released in the case of the $n+{}^3\text{He}$ channel to be divided in a 3-1 ratio among the final state products, so that the neutron energy is 2.45 MeV and the ${}^3\text{He}$ energy is 0.82 MeV. For the new reaction process leading to ${}^4\text{He}$ production in the Fleischmann-Pons experiment, much could be learned about how the reaction works from measurements of the alpha energy when created.

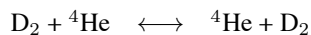
Unfortunately a direct measurement of this energy with a conventional silicon surface barrier detector is problematic, for a number of reasons. Such detectors are not compatible with a typical Fleischmann-Pons electrochemical cell, and a detector placed close to the cathode would likely impact the D/Pd loading, hindering excess heat production. This is not to say that it is impossible to develop such an experiment, but to date it has not been done.

A potential solution to this problem is to view the PdD itself (and also the D_2O) as an alpha detector [38]. Energetic alphas with sufficient energy have the potential to disintegrate nearby deuterons, which would lead to neutrons that could be detected outside of the electrochemical cell. Alphas with less energy can collide with deuterons, transferring energy, and these deuterons can undergo dd-fusion reactions with other deuterons if sufficient energy is transferred. A number of experiments were described in the literature in which excess power was observed in experiments monitored by neutron detectors. Excess power was observed to be uncorrelated with neutron signals in most cases, and anti-correlated in the case of experiments using a high-low current protocol. The upper limit was found to be on the order of 1 neutron per 100 joules of energy produced, which implies that the ${}^4\text{He}$ nucleus has a very low kinetic energy (under 10-20 keV) when created. Due to the large energy quantum involved (24 MeV), this implies a severe constraint on the reaction kinetics. In essence, there are no few-body incoherent reaction mechanisms that can be consistent.

The first step in the reaction process under discussion in this work is a phonon-mediated $D_2/{}^4\text{He}$ transition which is part of an excitation transfer process. The ${}^4\text{He}$ produced in such a process is born stationary, which is consistent with experiment.

16. Null reaction and ^4He

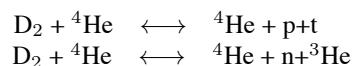
In our early work we recognized that the null reaction



involving the transfer of (24 MeV) excitation from one site to another was possible [40]. While this excitation transfer reaction as indicated here seems to involve a single D_2 molecule and single ^4He atom, the thought is that there would be a great many D_2 dideuterium species and a great many ^4He atoms interacting with common highly-excited THz vibrational mode (which would maximize the excitation transfer rate). As originally proposed it was thought that this kind of reaction was important theoretically, but would not be observable.

Recently it has become clear that the null reaction may be quite significant. If the null reaction occurs at a substantial rate, it would impact the radial D_2 wave function, potentially producing a substantial increase in the probability that the two deuterons are localized at the fermi scale. Although the Coulomb barrier is not affected, and the Gamow factor is not impacted, the null exchange process will lead to an increase in the probability amplitude at small separation due to the local transition $^4\text{He} \rightarrow \text{D}_2$. The proposal is that this effect is responsible for the observations of 3-body fusion reactions reported in [41], [42].

It has also become clear that if the null reaction occurs that one would expect associated occasional incoherent decays due to



It is possible that this ultimately may be the origin of the dd-fusion products in cold fusion experiments, and contribute to dd-fusion products in deuteron beam experiments at low energy [43].

This suggests that ^4He may be more important in experiments involved metal deuterides than we had been thinking in previous years. It may be that excess heat experiments in which the ease of starting up should be correlated with ^4He content. Similarly, ^4He content may be a parameter to monitor in experiments where low-level dd-fusion products are measured might.

In principle the same issues would apply in the case of HD and ^3He for NiH excess heat experiments. It may be that excess heat in the NiH system would start up more easily if some ^3He were present initially in the lattice. It may be that a 3-body reaction involving an HD to ^3He transition could be seen in a sample containing H, D and ^3He , with either H or D bombardment.

17. Vacancies

The issue of where the reactions occur has been of interest since the early days of cold fusion in 1989 [44], [45]. Deuterium occupies octahedral sites in bulk Pd, but the double occupancy of two deuterium atoms in an octahedral site is energetically unfavorable and does not lead to a smaller separation than in molecular D_2 . Our focus has been on monovacancies, in which the O-sites and T-sites energies are nearly degenerate, so that all of the 6 O-sites and 8 T-sites are potentially available for occupation. Due to an energy penalty for occupation of neighboring O-site and T-sites, we might expect there to be a constraint on the monovacancy loading where 9 or more D atoms are required in a vacancy before neighboring O-sites and T-sites are occupied. There is no advantage in tunneling for neighboring O-site and T-site occupation; instead, the thought is that this would allow for isomerization with dideuterium states (for which the D-D separation will likely be in the vicinity of 0.9-1.0 Angstroms, which is more separation than occurs in molecular D_2). The key issue here is not so much a potential increase in the tunneling probability, but instead that a significant coupling with extended vibrational modes seems possible.

In connection with this proposal is the observation of superabundant vacancies by Fukai and Okuma [46], [47]. The issue is that the vacancy formation energy is near 1.7 eV in bulk Pd, which means that it is energetically unfavorable for vacancies to form (and if large numbers of vacancies are produced by ion bombardment, most anneal spontaneously at room temperature within a day). If H or D atoms are in O-sites near a Pd atom, the binding energy for that atom is lowered, since the resulting monovacancy is a trap for H or D atoms. This leads to a reduction of the Pd removal energy by about 0.22 eV per H/D neighbor, so that PdH or PdD at high loading becomes thermodynamically unstable to decay to configurations with large numbers (25%) of vacancies. Since vacancy diffusion is slow at room temperature, in general one does not see massive vacancy formation in highly loaded PdD. In the experiments of Fukai and Okuma the associated lattice contraction at elevated temperature (700–800 C), where the vacancy diffusion rate is much faster, was observed in 2–3 hours.

In the Letts co-deposition protocol [48] excess heat is seen promptly when co-deposition is carried out at high current density (and low Pd ion concentration in the electrolyte), which is thought to lead to large numbers of monovacancies since new Pd layers are deposited under conditions where the D/Pd loading is high. This appears to be supportive of the notion that the active sites are monovacancies. In the Staker excess heat experiment [49] a protocol is used in which repeated loading of the Pd cathode through the miscibility gap, which would be expected to cause defects. From our perspective this would likely lead to a disordered lattice with a moderate (few per cent) monovacancy concentration, where Staker proposes that an order superabundant vacancy phase forms.

Note that Storms has advocated for micro-cracks as where the reactions occur [50]. In our view the sites next to the Pd surface in a micro-crack are similar to those in the interior of a monovacancy, so are candidates for reactions through the mechanism under discussion. The associated theoretical issue concerns how many sites are available. The associated experimental issue is the development and testing of well-characterized samples which have micro-cracks, and which have monovacancies.

There is the question of what happens in nano-Pd and nano-Ni particles. A proposal is that due to the considerable surface stress, and nearby surface from which vacancies can diffuse, that it is much easier for monovacancies to form in nano-particles. Possible support for this comes from x-ray measurements of PdD of [51] showing Fm3m symmetry (consistent with the Fukai superabundant vacancy phase), and neutron diffraction measurements of [52] also showing Fm3m symmetry. An interpretation of these measurements as indicating an ordered superabundant vacancy phase is possible, but this was not the conclusion in either paper.

18. D/Pd loading

In light of the discussion above an interpretation for the D/Pd maximum loading requirement of 0.95 in the SRI experiments may be connected with the stabilization of monovacancies. The idea is that if inadvertent co-deposition of Pd (or other metal atoms) occurs, then new layers formed would have many monovacancies. There are issues with this proposal. For example, Pd(+2) ions can be present in an acid electrolyte normally used for co-deposition, but not in base. Pd co-deposition in base either occurs much more slowly or not at all [53]. The surface of cathodes that have produced excess heat have been analyzed, and generally have all kinds of elements other than Pd on the surface. An alternate mechanism may be a combination of mechanical work and deuterium flux near the surface allows monovacancy formation at high D/Pd loading.

We would also expect a requirement that the D/Pd loading be sufficiently high to ensure sufficient occupation of the monovacancies. One thought was that this might be the origin of the early D/Pd threshold near 0.83. However, in light of observations of excess heat at much lower D/Pd loading by Storms [54], this interpretation needs revision. As the models suggest an analogy with laser gain, a proposal is that a high D/Pd loading of a relatively small number of monovacancies is needed to reach the threshold in the old SRI experiments, which occurred near 0.83. And that if there are many more monovacancies present, then a suitable occupation can be reached at a much lower D/Pd loading.

19. Phonons and flux

As the models for excess heat production under consideration are analogous to laser models, and light is needed initially (even if spontaneous) for amplification in a laser, phonons are needed initially for excess heat production. But where would THz phonons come from? In the early Fleischmann-Pons experiments it was noticed that excess heat was proportional to the deuterium flux above threshold [55] which suggested that the THz phonons resulting from D hopping in the lattice were driving the process. This is thought to be relevant for the Swartz fusor experiment, where the asymmetric anode arrangement would favor net deuterium flux into the cathode region close to the anode [56]. We have commented previously on the Letts 2-laser experiment [33], in which excess heat is stimulated by two laser beams when the difference frequency is matched to phonon modes with zero group velocity.

Of interest is how this might work in excess heat experiments with nano-particles [57]. One might expect excess heat to be triggered initially through a loading or deloading cycle, in which D/H generates THz phonons through diffusion (for which evidence is described in [57]). We recall this is how excess heat is stimulated in the Piantelli NiH experiment [58] with a macroscopic Ni bar in hydrogen gas. How D or H in a nano-particle is exchanged with the gas phase under quiescent conditions is of interest. It may be that once excess heat is initiated that the nuclear energy goes into the vibrations maintaining the excess heat process.

20. Discussion

The prospect of a clean small scale nuclear energy source has been in front of us at this point for more than 32 years. Meanwhile the CO₂ concentration in the atmosphere continues to rise and temperatures continue to rise. The lack of acceptance of the field by mainstream science persists, almost the same now as in 1989. And each year there are fewer researchers available that have a long experience in the field (Srinivasan will be sorely missed). The relatively recent interest on the part of industrial efforts has led to an increase in effort level, but an associated decrease in publicly available research results as we learn that industry does not seem interested in presenting or publishing.

Due to the hurdles faced by the field over the years progress in sorting out the associated physical mechanisms has been slow. In the end, how the excess heat process works is an experimental issue. Theory can provide some guidance, but experiments are needed to sort things out and to be sure of what is going on.

In view of the discussion above there are potentially a very large number of issues needing to be addressed and hopefully resolved. A theoretical construct exists which has the potential to provide a roadmap. Work continues on the models, and progress continues to be made (an examination of the discussion of the model in this work will reveal numerous additions and modifications from earlier versions [59], [26]). The biggest headache is that the theoretical picture is complicated with lots of parts. In the end there will need to be experimental tests of every piece (which is going to require elaboration of the models to provide quantitative predictions in each case, and new experiments that will allow quantitative comparison with theory).

So, what is to become of the field? My younger colleagues inform me that things are changing, and that a new generation is coming into their own that did not experience the trauma that was 1989. The words of Max Planck seem relevant in this connection [60]:

A new scientific truth does not triumph by convincing its opponents and making them see the light, but rather because its opponents eventually die and a new generation grows up that is familiar with it. . . . An important scientific innovation rarely makes its way by gradually winning over and converting its opponents: it rarely happens that Saul becomes Paul. What does happen is that its opponents gradually die out, and that the growing generation is familiarized with the ideas from the beginning: another instance of the fact that the future lies with the youth.

I recall a conversation that I had in 1989 with a well-known skeptic, who explained to me that it would be really nice if the excess heat in the Fleischmann-Pons experiment was real. The key problem, he said, was that he just didn't believe measurements done with isoperibolic calorimetry. If there were even one measurement done with a better calorimetric technique, such as flow calorimetry, then he assured me that he would become a believer in the effect. Some time later I informed him that some very nice positive excess heat results had been obtained at SRI in a flow calorimeter. He immediately became angry. He explained that the only way he would believe that energy had been produced would be if a commensurate number of neutrons were measured.

Unfortunately we do not have an abundance of young people in the field. And we do not have good teaching materials with which to pass on what we have learned to a next generation.

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