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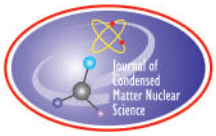

Research Article

Current Status of the Theory and Modeling Effort based on Fractionation

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Abstract

The theoretical problems associated with excess heat in the Fleischmann–Pons experiment were once viewed as insurmountable; nevertheless, some progress has been made in the past quarter century. Conceptually the problem can be split into one area involving new physics which is needed to address the microscopic physics of the reaction process; and a second area involving known physics which allows one to connect with the different practical issues involved in the experiments and observations. We review the ideas and progress first in the area of new physical mechanisms, in which models that describe the down-conversion of the large nuclear quantum allowing for coherent energy exchange of the nuclear energy into vibrational energy. These ideas provide a connection between excess heat experiments with Pd and Ni, with D or a mixture of H and D; experiments where tritium or low-level nuclear emission is seen, and other experiments with collimated X-ray emission. In the area of conventional physics mechanisms, we are interested in the basic physics of PdH and PdD; modeling cathode loading and understanding why some cathodes loaded very poorly in the early days; understanding active sites which we attribute to monovacancies in PdD and NiH; figuring out how active sites are created; triggering excess heat; and removing the helium made in the new reactions.
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1. Introduction

Ever since the announcement of excess heat in the Fleischmann–Pons experiment [1,2], theorists have generally struggled with the seemingly impossible theoretical ramifications. The majority of theorists in the scientific community generally regard the theoretical problems as impossible, concluding that the experiments which show anomalies simply must be in error. Even though a non-nuclear explanation appears to be ruled out by the absence of chemical products commensurate with the energy produced, and also by the very large estimates for the energy produced per unit volume, mass or atom; one still encounters a surprisingly wide variety of proposed explanations in terms of various chemical reactions, the water reaction, the release of stress energy, stored chemical energy, or mechanical energy. For those who accept that the origin of the energy must be nuclear, there is no agreement as to the nature of the microscopic

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mechanism; whether it be fusion, fission, some kind of neutron or nuclear fragment transfer, beta decay, more general nuclear disintegration, or perhaps some new kind of process involving exotic particles such as dark matter. Some regard the effect as implying some basic failure of modern physical theory, and have proposed alternatives to quantum mechanics, to field theory, and to the standard model. Quite a few theorists have focused on microscopic reaction mechanisms involving d+d and p+d fusion reactions; in this case a key issue has been finding some way around the Coulomb barrier in order to arrive at a predicted rate that is relevant to experiment.

2. Helium and Absence of Neutrons

^{4}He was observed in the gas phase by Bush, Miles and coworkers [3–6], and was found to be correlated with the energy produced [7]. Although there remains uncertainty in the energy produced per unit ^{4}He, a picture consistent with the majority of the measurements is one in which the energy release is due to the mass energy difference between D_2 and ^{4}He, and not all of the ^{4}He produced makes it into the gas phase unless steps are taken to scrub it out.

If we consider a microscopic reaction in which two deuterons react to make ^{4}He (independent of the many issues as to how such a reaction might happen), a natural question might be to ask how much of the 24 MeV reaction energy the alpha particle is born with. A knowledge of this energy could be helpful in clarifying the microscopic reaction mechanism. However, a direct measurement of the alpha energy seems problematic, as the introduction of a silicon surface barrier detector, or other conventional detector, would likely impact the experiment and inhibit excess heat production. It was proposed to view the PdD and D_2O present in the experiment as nuclear detectors [8,9] since energetic alphas would be expected to produce primary neutrons from deuteron disintegration, and secondary neutrons from inelastic collisions with deuterons (which could gain sufficient energy for subsequent dd-fusion reactions).

Experiments were found in which excess heat bursts were observed at the same time neutron measurements were carried out. In general there is no correlation between excess heat bursts and neutron emission, with an upper limit of about 1 neutron emitted per 100 J of energy produced. The conclusion from this analysis is that the alpha particles measured subsequently as ^{4}He have less than 0.1% of the 24 MeV reaction energy at birth, which rules out all plausible conventional Rutherford picture reactions. Energetic alphas if present would also lead to X-ray production [10], which is not seen in experiments. One could imagine the reaction energy somehow going into many making fast deuterons [11]; however, in such a scenario the energy would need to be distributed among more than 25,000 deuterons for the resulting neutron emission rate to be consistent with experiment [12].

3. Models Based on Down-conversion

For us the absence of energetic nuclear radiation in amounts commensurate with the energy produced was the most fundamental theoretical issue raised by the Fleischmann–Pons experiment, and was generally the focus of our early research effort. Our sense was that if the effect was real, then it implied the existence of a mechanism to down-convert a large nuclear quantum into a great many low energy quanta. In recent years we have made use of the term "fractionation" to indicate strongly enhanced down-conversion via the new mechanism reviewed in what follows. We began analyzing quite a few specific models of candidate systems seeking a down-conversion mechanism with no luck for many years. The first glimmer of success finally came in 2002 when we understood that down-conversion in the spin-boson model was limited by a destructive interference effect, and that the introduction of loss could remove this destructive interference [13]. The ideas, models, and progress on this problem have been reviewed several times over the years [14–17]. More general reviews are available in the form of video lectures based on IAP courses given at MIT in the past few years [18].

Although the early modeling focused on specific systems, it became evident that associated mechanisms could be studied more efficiently using "toy" models where specific transitions were abstracted into equivalent two-level

systems, and specific low energy condensed matter degrees of freedom (such as a vibrational mode) could be abstracted as an oscillator. The problem that results is one involving many two-level systems coupled to an oscillator. This problem is known in the literature, and falls under the general heading of a "spin-boson" model. We were interested in coherent energy exchange between the two-level systems and oscillator under conditions that the transition energy of the two-level systems is large, and the oscillator energy is small. Coherent energy exchange in the multi-photon regime of the spin-boson model is known, and we have contributed to the analysis of this problem and related issues [19–22].

The generalization of the model augmented with loss is complicated in general [17], and we focused initially on a restricted version where loss of infinite strength was modeled [23,24]. It had been found earlier that when the destructive interference effect was eliminated, the resulting coherent energy exchange rate was very fast, but depended relatively weakly on the details of the loss model. Hence we wanted to construct a simple but crude model which would give the maximum coherent energy exchange rate; this would be helpful in evaluating the model, and could provide a limit for more sophisticated models to be worked on later. With some work this model was analyzed systematically, and numerical and analytic results were established quantifying the coherent energy exchange rate and associated scaling laws [25–29]. The scaling in the limit where a very large quantum is down-converted to many small energy quanta was found to be algebraic, a gentle result which suggested a real possibility for the fractionation of an MeV quantum down to the eV range. Fractionation in this kind of model proceeds by the sequential exchange of many quanta one at a time, while overall coherence is maintained. The coherent energy exchange rate under conditions of fractionation is faster the stronger the individual coupling, the fewer small energy quanta involved, the more two-level systems present, and the stronger the oscillator excitation.

To connect with the Fleischmann–Pons experiment, interest focused on a transition from an initial D_2 state to a final ^{4}He state, with the energy going into a highly excited optical phonon mode. Since the Coulomb interaction inhibits tunneling, the resulting coupling matrix element is very small; hence this transition could never fractionate a large quantum. A solution was to transfer the excitation to other transitions strongly coupled to the oscillator, which would be able to fractionate the large quantum [30]. A key feature of this kind of model is that the associated coherent dynamics [31] ends up predicting a maximum coherent energy exchange rate linear in the coupling matrix, so that the Gamow factor associated with tunneling only comes in once [30]; as opposed to incoherent transitions where the Gamow factor comes into the rate quadratically. There has been much discussion over the past quarter century of how to avoid the Coulomb barrier [32], which in our view is impossible; instead, the impact of the Coulomb barrier on a coherent process is very much reduced. Estimates for the maximum reaction rate from this kind of model come out to be in the same range as experimental observations with no requirement for heroic screening effects.

Although the focus has been on the D_2/^{4}He transition for excess heat in connection with the Fleischmann–Pons experiment, it is clear that the same mathematical models would apply equivalently to HD/^{3}He transitions which are candidate reactions for light water experiments. Unimportant so far, but also similar is the HT/^{4}He transition. We have in the past proposed that a D_2/HT coherent reaction could be responsible for tritium production in PdD experiments. We worry about the D_2/n^{3}He channel which we fear might have been responsible for effects seen by Kevin Wolf [33]. In the case of light water systems, perhaps tritium can be produced by the beta decay of HD under conditions of incomplete fractionation.

Fractionation of the large quantum would be accomplished by nuclear transitions strongly coupled to the highly-excited oscillator. In the case of PdD systems, we contemplate the possibility that transitions in the deuteron may be responsible for optical phonon mode excitation, and transitions in the host Pd nuclei responsible in the case of acoustic mode operation. For Ni gas loaded systems we consider transitions in the host Ni nuclei as candidates for the fractionation. Note that fractionation in the host lattice nuclei has the potential to lead to elemental anomalies [34,35] through a coherent disintegration process [36].

4. Interaction and Collimated X-ray Emission in the Karabut Experiment

Once a mechanism was found that could down-convert a large nuclear quantum into a large number of smaller quanta, it was thought that a theory for excess heat in the Fleischmann–Pons experiment would quickly follow. Unfortunately, there were more difficulties. The rate at which excess heat was produced in experiment seemed consistent with the maximum rate predicted in the donor and receiver model [30] if the associated MeV-level quantum were fractionated; and it was clear that the scaling laws were sufficiently gentle that an MeV-level quantum could be fractionated down to quantum below an eV; however, try as we might we were unable for years to identify a specific transition capable of the required fractionation. The basic problem was that the weak second-order coupling leading to phonon exchange in connection with a nuclear transition was far too weak to mediate the needed down-conversion.

Progress on this problem next came from an unexpected quarter. Since there are not energetic nuclear products commensurate with the energy produced in the Fleischmann–Pons experiment, it does not seem possible to prove the reaction mechanism under consideration in this review by direct measurements. On the other hand, if the down-conversion mechanism is involved, we should be able to study it in isolation in some other application. We pursued the design of such an experiment which focused on up-converting many low energy vibrational quanta to produce a large quantum needed for nuclear excitation. The led to consideration of an experiment focusing on nuclear excitation in ^{201}Hg, where the 1565 eV transition is the lowest among stable nuclei starting from the ground state [15]. Subsequently came the interpretation of collimated X-ray emission near 1.5 keV in the Karabut experiment [37–39] as arising from the up-conversion of vibrational energy leading to phased array collimated emission from impurity Hg nuclei as contaminants on the surface.

It became clear that we should be able to apply the new model to analyze the Karabut experiment; and when done initially we concluded that the weak second-order interactions which we had been working with were just not strong enough to produce the effects observed by Karabut. Whatever interaction produced nuclear excitation and exchanged phonons needed to be orders of magnitude stronger. Ultimately this led to the identification of the first-order interaction Hamiltonian from a relativistic description of the nuclei in the lattice [40,41] as the only possible interaction capable of producing the requisite fractionation power. Still, there remained problems in making a connection between theory and experiment. The basic picture being contemplated for the Karabut experiment involved vibrations in the small metal cathode as responsible for the up-conversion; however, we were not able to get quantitative agreement between theory and experiment [42,43]. Incredibly, within this picture the coupling seemed still to be too weak. The basic model was re-analyzed, and a new regime of the model was found where the coupling was much stronger [44], which seemed to provide a resolution.

Meanwhile, tests for X-ray emission and charge emission from a vibrating copper foil were ongoing at SRI and later at MIT [45,46]. These experiments did not produce X-ray emission under conditions where one might have expected if the above interpretation of the Karabut experiment were correct. In the anomalous regime of the model there could be sufficient up-conversion strength in the copper foil, as long as sufficiently strong transitions were present to get into the anomalous regime in the first place. For a copper foil, the lowest energy excited states are above 500 keV (too high to be useful), so the only candidate transitions strong enough to reach the anomalous regime in principle are those connecting to negative energy states. Hence, the tentative interpretation of a negative result from the SRI and MIT experiments with copper is that transitions with negative energy states do not mediate up-conversion and down-conversion [47]. This suggested a revised interpretation of the Karabut experiment, where MHz vibrations in the much more massive vacuum chamber was responsible for the up-conversion, with the 14.4 keV transition in ^{57}Fe mediating the conversion. Such an interpretation brings into alignment the water jet experiment of Kornilova et al. [48] where collimated X-rays are reported observed from a nearby steel plate.

We note the possibility of up-conversion to produce coherent disintegration [36] in connection with the experiments of Carpinteri and co-workers [49]. If correct, this suggests the possibility of the up-conversion of vibrational energy to

produce disintegration in some high-Z element (such as lead) for excess heat production by cold fission.

So far there have been calculations of two matrix elements based on the new phonon-exchange interaction. One is for coupling with deuterons in the lattice [50]; which leads to selection rules which appear to be consistent with magnetic field effects having a substantial effect on excess heat production in PdD systems [51,52]. The other is for phonon exchange in $D_2/^4He$ transitions [53], which results in a matrix element sufficiently large to be relevant to excess heat production experiments [54].

5. Physics of PdD and D/Pd Loading

Deuterium is loaded into palladium electrochemically in the Fleischmann–Pons experiment, so to model the Fleischmann–Pons experiment we need to understand some of the associated (conventional) physics. One can find reviews of metal hydrides in general and palladium hydride in particular in the literature [55–61]. Over the years people have used a variety of theoretical methods to model hydrogen in Pd, starting with the important analysis of [62]. We pursued density functional calculations [63,64] in an effort to understand the phase diagram from first principles; from this effort we understood that even though the accuracy of such methods is very good, one needs better accuracy to model the phase diagram. From our perspective, the best approach to develop the models needed is an empirical one. We recently made use of empirical models including both octahedral and tetrathedral site occupation to compare against experimental data for α-phase PdH_x and PdD_x, with the result that it is possible to estimate quantitatively model parameters for both the O-site and T-site energies and other model parameters [65]. A similar model was used to compare with experiment in the case of high loading [66], and once again we were able to extract estimates for the model parameters. The resulting models are useful in describing PdH and PdD in the β-phase up to higher than stoichiometric loading.

Hydrogen and deuterium evolution reaction models are used to model the electrochemical loading of the Pd surface and bulk. We were interested in these models in order to understand why there was so much difficulty in obtaining high D/Pd ratios in the early years following the announcement of cold fusion in 1989. We focused on a subset of the reactions in the Volmer-Tafel regime, and noticed that there was a big variation in the resulting model parameters when different specific experiments were considered. Our conclusion was that it was likely that the rate of internal D_2 leaks in the different cathodes varied by as much as three orders of magnitude [67,68].

For modeling the solubility and also the loading of D and H in Pd we need a good model for the fugacity of H_2 and D_2 gas, especially under conditions of high loading. We examined different empirical models that have been developed over the years [69] and concluded that the best available model at this point is a recent one due to Joubert [70]. In this study we were also concerned with the difference between the fugacity of H_2 and the fugacity of D_2, concluding that the small difference between them was not accurately dealt with over the full range currently by any set of models.

6. Monovacancies and Co-deposition

It was recognized early on that getting two deuterons close together in PdD was problematic due to the high background electron density due to the Pd [71]. It was also noted early on that the repulsion between hydrogen or deuterium atoms was reduced in vacancies [72] since the background electron density is reduced. Another early paper focuses on the possibility of close encounters between deuterium atoms in a monovacancy with 6–9 deuterium atoms [73].

Sigma-bonded dihydrogen complexes involve the bonding of molecular H_2 as a molecule; they were discovered in the 1980s [74], and have been the focus of much subsequent research. For many years we have been interested in the possibility of dideuterium formation in a monovacancy in PdD [75–77]. The basic idea is that deuterons would be in close proximity in such a configuration, and the high background electron density is helpful for screening (enhanced screening is thought to play a role in low-energy deuteron beam experiments [78]). A dihydrogen complex similar to what we proposed has been found on the surface of highly loaded PdH [79].

In the past few months a modified version of this scenario has become of interest to us. We note that the O-site and T-site states around a monovacancy in Pd and Ni are nearly degenerate [81,80], so that when H or D atoms occupy a monovacancy they might occupy either O-sites or T-sites democratically until repulsion effects become important. Hence with an occupation of six deuterium atoms, a stable configuration could be where all the O-sites are occupied; and with eight deuterium atoms, all the T-sites could be occupied; in both cases there are no O–T nearest neighbors. When deuterium atoms occupy neighboring octahedral and tetrahedral sites, there is a repulsion energy [82] on the order of 150 meV. Current thinking is that the deuterium chemical potential becomes sufficiently high above a bulk D/Pd loading of 0.83 for neighboring O–T occupation in monovacancies, where tunneling might be enhanced by screening due to background electrons. This kind of configuration would be similar in Pd and Ni monovacancies, and probably for other FCC lattices as well. It seems likely something similar can happen in vacancies in BCC metal deuterides.

Probably a large number of monovacancies is required for excess heat production in the Fleischmann–Pons experiment, so the issue of how they are made becomes relevant. The vacancy formation energy in Pd is near 1.6 eV, which implies a low vacancy population in thermal equilibrium. However, the vacancy formation energy is decreased for every neighboring H or D atom, and at sufficiently high loading the vacancies become favored in thermal equilibrium. This was exploited by Fukai and Okuma [83,84] who created vacancy phase PdH and NiH which had 25% host metal atom vacancies. High loading is known to be important in the Fleischmann–Pons experiment. We have interpreted the requirement that Pd cathodes must reach a D/Pd loading of 0.95 [85] for excess heat to be seen as consistent with where vacancies are stabilized near room temperature. The problem is that even if stabilized, vacancy diffusion is sufficiently slow at room temperature that one would expect negligible formation of superabundant vacancies in a Fleischmann–Pons cell due to diffusion alone. We proposed that superabundant vacancy formation should occur in new material created through co-deposition, so that the monovacancy active sites are near the surface. We interpreted the early Szpak co-deposition experiment [86] as producing excess heat with no need for an extended loading period due to superabundant vacancy creation due to co-deposition under conditions of high loading so that vacancies are thermodynamically preferred. Support for this point of view comes from the Letts high-current density co-deposition protocol [87], which was confirmed by Miles [88]. This approach was exploited for cathode preparation in the cryogenic experiments done recently at SRI [89,90]. Co-deposition at lower current density would be expected to make fewer vacancies, resulting in insufficient active sites for sufficient gain for excess heat production, but which result in low-level nuclear emissions [91].

In the case of the NiH gas loading experiments of the Piantelli group [92,93] the question might be asked as to how monovacancies might be created since hydrogen solubility is so low in bulk Ni. In this case, if the H/Ni ratio is sufficiently high so that the loading is in the mixed phase region of the miscibility gap, then H is present in regions of α-phase and β-phase NiH_x. This is interesting as there is the possibility that the hydrogen density in the regions of clumped β-phase material to be sufficiently high to stabilize the vacancies. At the elevated temperature of the Piantelli experiment, there is the possibility of some vacancy diffusion. This potentially provides a mechanism for the development of increasing levels of vacancies with cycles of H loading and de-loading near the surface. If the Ni contains impurities, there is the possibility that the sites near the impurities will look attractive the the H, and also that the vacancy formation energy for the vacancy will start out a bit lower than for Ni. This may be relevant to the claims made for the recent experiment of Parkhomov (discussed at ICCF19), which involves impure Ni and quite high temperature.

In light of the comments above, it seems clear that the Pd and Ni structures produced in experiments to make superabundant vacancies would be attractive candidates for experimentation, since the monovacancy concentration in maximized. We are not aware of such experiments having been carried out at this point, even though we have advocated this approach for two decades.

7. Other Issues

Suppose that a candidate material has been produced with a large number and density of monovacancies, and that the sample has been loaded with whatever mixture of D and/or H is desired. Of interest then is to stimulate the sample in order to trigger excess heat production. Within the approach under discussion there are some theoretical issues to be considered. Vibrations in the lattice involve a large number of modes; to maximize the coherent dynamics of the models we are interested in an ideal system in which a single highly-excited vibrational mode is present, preferably one which interacts with D_2 or HD equivalently at all of the active sites. In PdD the Γ-point optical phonon mode works this way, and also has the added advantage that the group velocity is zero, so that vibrational energy generated at the Γ-point will remain largely localized in space. This leads us to consider Letts' two-laser experiment [94,97], in which two visible lasers tuned so that the difference frequency is in the THz range is scanned. Maximum excess heat is found at three frequencies; the lowest near 8.2 THz, identified with the optical phonon modes near the Γ-point; a second near 15.1 THz, identified with optical phonon modes near the L-point (also where the group velocity is zero); and a third near 20.8 THz. The latter peak does not have a confirmed identification; we have speculated that it might be due to the L-point associated with H impurities. We note that a confirmation of this experiment has not yet been obtained. Of interest in these experiments is that once excess heat has been stimulated in the two laser experiment, it is observed to remain on, or to decay slowly following shutting off the lasers. An interpretation of this effect is that the two lasers introduce an initial excitation in the optical phonon mode locally causing nuclear energy from the coherent process to be drawn into the excited modes (analogous to what happens in a laser); probably the area where the reactions occur expands over several hour time scale. When the lasers are turned off, the thought is that the nuclear energy continues supporting excitation of the vibrational modes, providing indirect support for the conjecture that nuclear energy is down-converted in this experiment. Direct support for the conjecture would come if large amplitude optical phonon vibrations were seen directly in these experiments, either with optical Raman type measurements, or with inelastic neutron scattering.

This suggests that triggering excess heat could be accomplished simply by providing excitation in high frequency vibrational modes, which could be done through interstitial hydrogen or deuterium diffusion, ion bombardment, electrical stimulation, or electromagnetic stimulation (excess heat has been observed under conditions which could be interpreted involving THz vibrational excitation by each of these mechanisms). In the event that the vibrational mode loss is low, as in the two-laser experiment, it may be that the vibrational energy can be maintained so that no further stimulation is needed. If the group velocity is high so that the vibrational energy leaves the area where the monovacancy active sites are, or if the vibrational modes involves some complicates superposition of local plane wave modes excited by interstitial diffusion, then one can imagine that the loss of vibrational energy is sufficiently high that the coherent process can only be maintained by continued vibrational excitation. In the first case, we might identify sustained excess heat production with what has been termed "Mode B" operation (in which the excess heat in a Fleischmann–Pons cell is nearly independent of the current density once initiated), while the second case would correspond with the more typical "Mode A" operation (where the excess heat is strongly correlated with the electrochemical current density).

If the system is working well, and D_2 is converted to ^{4}He (which is born stationary with all of the reaction energy going into vibrations), then one might expect ^{4}He accumulation in the monovacancies. This helium accumulation would be expected to clog up the monovacancy active sites, limiting the rate of excess power production. This picture could account for the observation that power levels seem to increase as the temperature gets higher, as helium diffusion away from the active sites is more efficient as the temperature increases. In some experiments the temperature dependence of the excess power is consistent with the helium diffusion coefficient [7]. One could imagine conditions under which the bottleneck might be the initial removal of the helium from the monovacancy which would involve a

higher barrier energy [97]. We have proposed that the issue of helium removal may account for the advantage of nano-scale excess heat systems, such as described in [98].

According to the models, the ability of an equivalent lossy spin-boson system to fractionate a large quantum depends on the microscopic coupling strength, as well as other factors including the Dicke number [27], which can be larger if there are more two-level systems. There are a number of implications for this. One is that the HD/^{3}He transition is one that is "easier" to work with since the large quantum is 5.5 MeV which is less than the 23.9 MeV of the D_2/^{4}He transition. One could imagine a cold fusion system which runs with the D_2/^{4}He transition when it works well, but if it were running less well so to be unable to fractionate the large 24 MeV quantum, one could imagine it switching over to the D_2/HT transition. One could also contemplate a frustrated version of the system only able to fractionate a much smaller quantum, which perhaps would lead to low-level nuclear emissions as seen in the SPAWAR experiments [91].

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