Fleischmann-Pons effect studies

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Introduction

The Fleischmann-Pons experiment has been the source of much controversy since first announced in 1989. In this experiment, a Pd electrode is loaded with deuterium electrochemically in a 0.1 M LiOD D_2O electrolyte, and sporadic increases in the cell temperature are observed. The amount of energy that it would take to produce a single thermal increase is very large. For example, in one example described in [1] the energy produced was 4 MJ from a cathode with a volume of 0.157 cm^3; if that same volume were replaced by TNT and detonated, the chemical energy produced would be about 1.2 kJ. As a result, Fleischmann conjectured that the energy was nuclear in origin.

The basic problem with such a conjecture is that nuclear reactions have been studied for close to a century at this point, and energy produced in nuclear reactions comes out in energetic particles. This was understood early on as a consequence of energy and momentum conservation in the reacting system when analyzed as an isolated system. In the Fleischmann-Pons experiment, no commensurate energetic particles are seen corresponding to the energy produced. As a result, either the energy must be due to experimental error, or else some new kind of physical process is responsible.

Following the initial announcement of 1989, the consensus among the scientific community was that the experiments were in error, and the thermal effect claimed was an artifact. The absence of energetic particles played no small role in the arguments made at that time, and one can find upper limits being claimed on the nuclear energy produced from upper limits on energetic particles [2]. Local conservation of energy and momentum in nuclear reactions remains a foundation of nuclear physics, and the claims of the two electrochemists (Fleischmann and Pons) was insufficient to cause the physics community to reconsider things.

The rejection of the claims of Fleischmann and Pons was a convenient way to re-establish self-consistency and order. Local energy and momentum conservation remains a foundation of nuclear physics, the energy from a nuclear reaction is expressed as energetic particles, and Fleischmann and Pons could safely be cast out of the scientific community with their scientific reputations destroyed. The only problem with this tidy solution is that the thermal anomalies claimed by Fleischmann and Pons continue to be seen in the laboratory in a very large number of experiments carried out by those who have continued to study the problem. There was a time when an important experimental result would make a difference; and if the result was not in agreement with theory, then efforts would be made to craft a better theory. In this day and age, seemingly everything there is to know about nuclear physics is already known, so that it is much more productive to discredit everyone associated with the Fleischmann-Pons experiment, than to support efforts to clarify the new science or to consider the implications of the result.

Over the years there has been progress on the problem, in a variety of areas, as we will summarize below.

Energy gain

In the early days, it was argued repeatedly that the energy output associated with the Fleischmann-Pons experiment was small compared to the total energy input over the history of the experiment. In a typical experiment at that time, a month long charging time seemed to be required prior to the observation of excess power events. The energy produced in a few hour long event was weighed against the energy input over a much longer period. The fact that the energy output in one of these isolated "burst" events could be 100-1000 times more than the energy available from the volume equivalent of TNT was deemed irrelevant. So, in one of the best early SRI results where a clear excess power event was observed in an accurate flow calorimeter, with an excess power signal above the noise by several 10s of sigma, the critics were quick to argue that the energy gain over the entire duration of the run was about 3%, so that it could conveniently be dismissed as within experimental error. The associated problem of how so much power could be produced sustained over many hours could be ignored using this kind of argument.

In more recent experiments, Energetics reported at ICCF10 the observation of an energy gain of 6.72 in a glow discharge experiment with a flow calorimeter [3]. Following this, a collaboration was formed in which Energetics teamed up with ENEA Frascati, SRI and NRL to produce the leading scientific collaboration in the field. At ICCF11, Energetics reported the observation of an electrochemical cell which in which the excess power was observed to rise to about 50 times the input power, and the energy gain integrated over the experiment history was 33 [4]. By now, this very large energy gain has been seen in roughly 5 cells, while the energy gain from more typical experiments are in the range of 100-300%.

In some experiments, the excess power persists after the electrochemical current has been terminated, and effect termed "heat after death" by Fleischmann and Pons who first reported it. The associated power gain in such experiments is infinite. Swartz has recently reported a correlation between the excess power and the square of the open-circuit voltage in his experiments.

At this point there has been a large number (several hundred) of observations of the Fleischmann-Pons excess heat effect reported, in a variety of different kinds of experiments, using a variety of different calorimetric techniques [5,6].

One would think that the question of the existence of the Fleischmann-Pons excess power effect itself should no longer be in any doubt. Yet in a recent interview on 60 Minutes, Dick Garwin expressed the (mainstream) opinion that he thinks that everyone who has measured excess heat in a Fleischmann-Pons experiment has simply made a mistake. We have heard that all proposals submitted to the recent DoE ARPA-E energy initiative that were for research on the Fleischmann-Pons effect were rejected unread.

Helium

Some critics argued in 1989 that the Fleischmann-Pons experiment was a perpetual motion machine in the sense that the energy was coming out of nowhere (which would be the inevitable conclusion if the energy was not of chemical or nuclear origin). Fleischmann's early conjecture was that somehow deuteron-deuteron fusion was responsible, indicative of a point of view that the experiment started with a nuclear fuel, consuming it in some new way, and ultimately leaving an ash. Unfortunately, if there are not energetic nuclear products, then it is hard in general to figure out what the ash is since there are so few products produced for a given amount of energy.

Fleischmann and Pons looked for tritium and helium, but did not find either in amounts commensurate with the energy produced. Initially, there was no input from experiment giving direction as to what elements or isotopes to look for, resulting in exhaustive and expensive searches for isotopic anomalies among the Pd isotopes, Li isotopes, and elemental assays of the cathodes. A variety of elemental and isotopic anomalies were seen, and this resulted in much controversy as to their origin. The situation was clarified later on by the observations of Miles and Bush of ^{4}He in the gas phase, correlated with the energy produced. Such experiments are not easy, and they are very expensive (relative to a field which historically has been short of resources). Confirmations of this result were reported by a number of groups, including Gozzi et al, SRI, and ENEA Frascati.

By itself, the observation of ^{4}He correlated with excess energy is interesting, but is not particularly helpful since there are a very large number of candidate nuclear processes which the theorists have suggested which involve ^{4}He in some way as a product. Much more important is the reaction energy along with the observation of helium. Since the Fleischmann-Pons experiment requires deuterium, and since helium seems to be a product, the simplest conjecture might be that somehow two deuterons interact to make ^{4}He. If so, then one would expect that the mass difference should be the reaction energy

$$M_D c^2 + M_D c^2 - M_{4He} c^2 = 23.86 \text{ MeV}$$

Even if it is unknown how this might work, it provides a candidate reaction energy which can be used as a measure against the experimental observations. This was done in the work of Miles and Bush, who found a wide scatter in their experiments. Later experiments by Gozzi and coworkers found helium correlated with the energy, but with the amount of helium different for the different heat bursts that were seen. The amount of helium seemed in some experiments to be close to the amount expected from the mass difference, and less in others. Although Gozzi offered no interpretation, the interpretation which was obvious to some of those listening was that some of the helium was retained in the cathode. If the excess heat was produced near the cathode surface, then one would expect some of the helium to diffuse out and be measured, but perhaps not all.

Subsequent experiments seemed to be consistent with this point of view, with the amount of helium measured being between 50% and 70% of the amount expected based on the mass difference. However, to be sure, there needed to be new experiments in which an effort was made to scrub all of the helium out of the near surface region. This was done in the SRI M4 experiment, and to within experimental error the amount of ^{4}He which was seen was consistent with the energy produced taking the reaction energy to be the mass energy difference [7]. For years, this result stood alone as the only one of its kind. Recently, a second such experiment was reported (ENEA Frascati Laser-3), where an effort was made to scrub the retained helium out, and it gave similarly helium correlated with the energy consistent with 24 MeV per He atom [8].

Where does the energy go?

Based on the discussion above, the experiments seem to suggest that deuterons react in some new kind of way to make ^{4}He, with the reaction energy going somewhere other than into energetic particles. The energy is seen after it thermalizes, but as yet there are few results that tell us where the energy goes prior to thermalization.

There are spirited arguments among scientists interested in this problem as to where the energy goes. One group steadfastly maintains that the energy has to be kinetic, and that it just hasn't been looked for yet. However, there have been a great many experiments in which efforts were made to detect radiation, and none of which established any correlation between the energy produced and any signal. An anticorrelation was found between excess heat and neutron production in experiments with a high-low current protocol, suggesting that cathodes which produced excess heat at high current density did not produce neutrons at high current density, but instead did produce low-level neutrons at low current density. The amount of neutrons in this anticorrelation were more than 10 orders of magnitude less that would be required to account for energy production had they occurred in the high current density phase. At present it is clear that we need new experiments which put concrete upper limits on the different kinds of energetic particles which have been suggested as candidates.

Previously, we had argued that the absence of Pd K-shell x-rays could be used to develop an upper limit on the alpha energy, but this argument has been criticized recently based on the difficulty of energy transfer between a "heavy" alpha particle and "light" electron. Hence, new arguments are needed which address the upper limit on the alpha particle energy in these experiments.

Perhaps the most relevant experiment in this regard comes from a different effect which is thought by some to be closely related. Significant amounts of tritium have been observed in Fleischmann-Pons and related experiments. Kevin Wolf was interested in the question of the triton energy when created, which could be studied using neutron spectroscopy. In a metal deuteride, an energetic triton will react with a deuteron to produce a 14 MeV neutron, which can be seen with an energy-resolving neutron detector. Wolf teamed up with Tom Claytor, who was studying tritium production, and measurements were made assaying for neutrons in experiments where significant tritium was produced. In these experiments, there was no evidence for any 14 MeV neutron emission. From the associated upper limit, Wolf deduced that the maximum triton energy possible at birth was about 8 keV. This can be compared with the 1 MeV energy of a triton formed in the d(d,p)t reaction.

Since tritium is essentially stationary when created, it seemed plausible that ^{4}He in the excess heat effect is also born essentially stationary.

Nevertheless, such arguments are not uniformly accepted. In recent presentations different theorists have advocated for exotic reaction mechanisms which would result in energetic alpha emission between 12 and 24 MeV at levels commensurate with the energy produced. This seems to us to be fundamentally inconsistent with earlier searches for radiation, and with the constraints imposed by the helium gas experiments. For example, if the helium is born energetic, then how can 70% of the total amount be seen in the gas phase, given that the stopping distance is very much larger than the diffusion distance?

Two-laser experiments

There have been numerous suggestions made over the years as to where the energy might go if not into energetic particles. Essentially every available degree of freedom has been suggested as a candidate in one model or another. Our initial proposals concerned optical phonon modes; in Preparata's model the energy was proposed to go into plasmon modes. However, conjectures and models are easily produced, but we need relevant experimental results to understand which is right.

Some indirect indication is available in single laser experiments [9]. Letts and Cravens presented results from their experiments at ICCF10 which indicated that a Fleischmann-Pons cathode could be stimulated to produce an excess heat event by a red laser [10]. Lasers operating between 1 mW and 30 mW and focused to a spot about 1 mm in diameter were seen to stimulate excess heat events that involved much higher excess power levels. If one follows the intuitive argument from laser physics that an amplifier will want to add energy to modes that already have energy, then these experiments could be viewed as providing indirect evidence that the nuclear energy was somehow ending up in the plasmon modes prior to thermalization. Since the effect is polarization-sensitive, the statement is probably a stronger one: that the energy is going into longitudinal plasmon modes (the ENEA Frascati group showed that p-polarized light was required to produce excess heat, and that this polarization couples to longitudinal plasmon modes [8]). The plasmon modes are very lossy, so that it is not easy to sustain excitation in them. In single laser experiments reported by McKubre et al, the excess heat events were seen to turn on following the laser being turned on, and turn off following the laser being turned off.

Letts and Cravens recently demonstrated a two-laser effect that is sensitive to the difference frequency of the lasers [11]. In these experiments, it was observed that at certain difference frequencies it was easy to obtain an excess heat response, while at other difference frequencies the cell showed little or no response. Over the course of a very large number of experiments done painstakingly during two calendar years, data was accumulated to produce a spectrum. One sees in the spectrum below a response at three different frequencies in the THz regime.

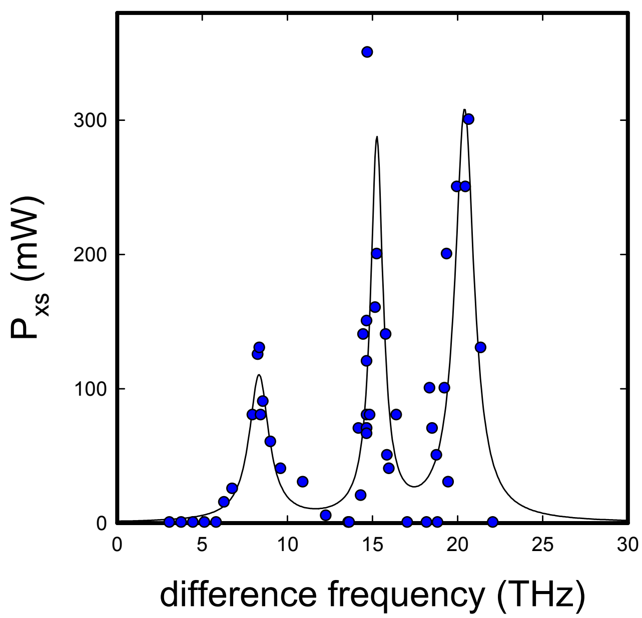

Figure 1: Excess power as a function of difference frequency in the two-laser experiments of Letts and Cravens.

Prior to the experiment, we suggested that a response might be observed near 8 THz and 16 THz, which correspond to the zero-group velocity points of the compressional optical phonon modes of PdD. The dispersion relation from a Born-von Karman model fitted to coherent neutron scattering data for $PdD_{0.63}$ [13] is shown in Figure 2. One sees that the phonon frequency at the L-point is computed to be around 16.6 THz, which seems to be in the vicinity of the excess power response peak at 15.3 THz. In the coherent neutron scattering data, the data points at the L-point in one experiment ([13], $PdD_{0.63}$) show up near 16 THz, while in another ([14], $PdD_{0.78}$) they are closer to 15 THz. The model frequency at the Γ-point is about 9.0 THz, in agreement with the coherent neutron scattering experiments, but there are incoherent neutron scattering experiments that show a peak closer to 8 THz. We consider the excess power response near 8.3 THz as being connected with the Γ-point compressional optical phonons.

However, there remains the question of the signal at 20.4 THz, which lies above all of the PdD optical phonon modes. In this case, we conjecture that it is due to H contamination in PdD. Due to the higher solubility of H in Pd as compared to D, the H/D ratio in Pd at about 10 times the ratio in the electrolyte. We have computed the phonon spectrum in the case of PdD with a 1 in 8 replacement of D with H in an ordered lattice as an analog to the non-ordered mixed problem, and we find that the L-point branch associated with the H occurs at 19.7 THz. This supports the identification of the 20.4 THz signal with L-point equivalent H contamination.

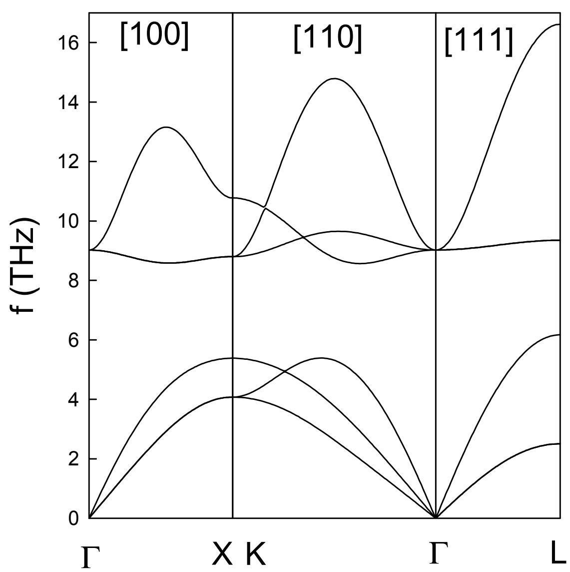

Figure 2: Born-von Karman computation of the phonon mode structure of PdD.

In some of these experiments it was observed that the excess heat would often persist after the termination of the two-laser stimulation. This suggests a picture in which the two-laser stimulation initiates an excess heat event, with the nuclear energy finding its way into the longitudinal optical phonon modes with zero group velocity, which are then excited to very high levels. When the lasers turn off, there is no sudden change seen in the level of excess power in many experiments. If we think of the high mode excitation as being the important aspect of the experiment, then we view the two lasers initially stimulating plasmon modes, which are then enhanced by the nuclear energy. But we know from earlier work that these modes are hard to sustain. But in these experiments the effect is sensitive to the beat frequency, which suggests that the plasmon modes (which are hybridized with optical phonon modes) become strong enough to interact nonlinearly, generating internally the difference frequency. Then, once the optical phonon mode excitation is sufficiently strong to draw the nuclear energy by itself, then it becomes self-sustaining. When the laser excitation is removed, the initial plasmon modes quickly lose their excitation, but the optical phonon mode response continues.

Discussion

We have focused our attention on some of the basic issues involved in excess heat in the Fleischmann-Pons effect. In spite of the continued spirited denial of the existence of the effect by Dick Garwin, and by most other physicists, the effects continue to show up in the lab. We are able to learn more about the effect by studying the experiments and the results from observations. The energy produced is real; it is a reproducible effect; and it is seen at levels orders of magnitude greater than can be accounted for by chemical processes. The energy is correlated with ^{4}He production, and the energy produced per ^{4}He atom observed is about 24 MeV to within about 10% as a (still preliminary) experimental result. The two-laser experiments show a response at difference frequencies in the THz region, with resonances which seem to correlate with zero-group velocity points in the compressional optical phonon modes of PdD, and the L-point mode due to H contamination. This provides strong indirect evidence that in these experiments the nuclear energy is showing up in these optical phonon modes and sustaining them when the lasers are turned off. A direct measurement of these optical phonon mode amplitudes would provide direct evidence, but such measurements have not yet been attempted. Evidence from single laser experiments provides indirect evidence that hybrid plasmon-optical phonon modes can receive the nuclear energy. We tend to associate localized hot-spot melting and elemental anomalies with the nuclear energy going into acoustic modes.

Although not discussed in this article, we have made much progress on the modeling during the past year. Although we have been looking at the same basic mathematical models for a number of years, during this past year we seem to have looked at them with new eyes, since they seem now to mean much more than before. We are now able to derive the coupled models for two-level systems and an oscillator that we have described in previous reports. The coupling matrix elements for excitation transfer now can be interpreted as being composed of a product of three terms: the strong force matrix elements between D_2 and ^4He; a Mossbauer matrix element restricting energy exchange with thermal modes; and a Franck-Condon factor for non-zero phonon exchange with the highly excited mode. We can explain the dependence of excess heat in the SRI experiments on loading now with a simple argument based on the thermodynamics of D_2 formation in monovacancies in PdD. We have previously discussed the high 0.95 loading requirement in terms of vacancy stabilization in PdD. Recently we have developed a new model for the electrochemical kinetics for the deuterium evolution reaction which provides a much better account of the observations of loading as a function of current density.

We are in the process of developing a simulation model for the excess heat effect in the Fleischmann-Pons experiment and the SRI version of it. In the latter case, there is a fair amount of experimental data available which we will be able to test our model.

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