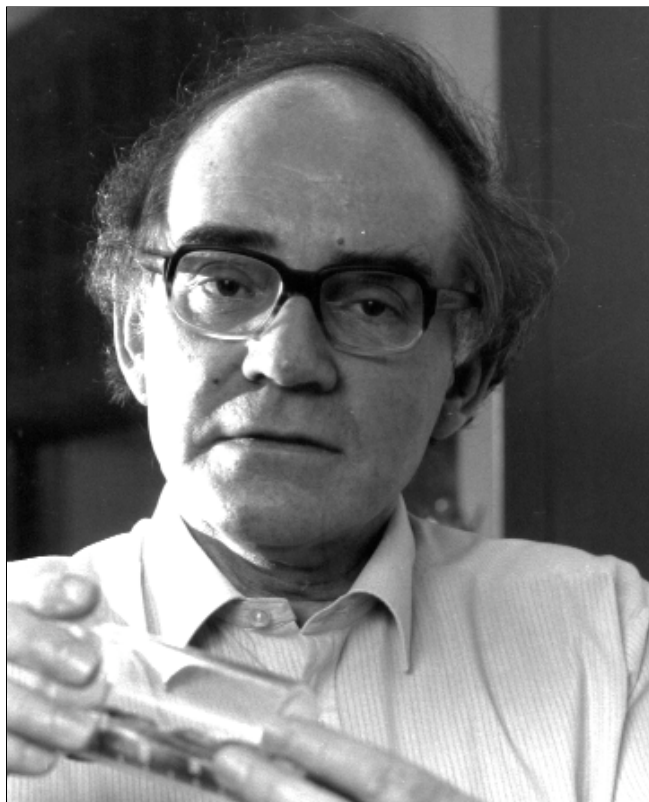


Nuclear Reactions in the Pd/D System: The Pre-History and History of Our Early Research

by Martin Fleischmann, FRS



Background

In the 1960's I became convinced that the behavior of ions in solution could only be explained in terms of Quantum Electrodynamics (Q.E.D.), rather than in terms of Classical Mechanics (C.M.), or Quantum Mechanics (Q.M.). I will not outline here the tortuous path which led to this conclusion; instead, I will use an illustration of the nature of the problem.

The generally accepted model of electrolyte solutions, the Debye-Hückel Theory, is based on the calculation of the energy of the systems due to the electrostatic interaction of the ions, Figure 1, *i.e.* on C.M. with some allowance for the fact that we are dealing with a many-body problem. In point of fact, the ions are not at rest and it has been known since the last cen-

tury that ions move independently of each other at high dilutions (Kohlrausch). Moreover, the dominant motion of the ions is attributed to Brownian movements (Einstein) so that the ions must accelerate and decelerate, Figure 2. The concomitant radiation (Maxwell) should therefore cool the electrolyte solutions to absolute zero at which point the Debye-Hückel Theory would become valid. We therefore need to ask ourselves: why do the solutions not cool spontaneously? The answer is that the electrostatic model, when combined with Brownian motion, violates the Second Law of Thermodynamics (see Appendix A). In point of fact, the motion of the ions must be rigorously quantized and the correct theoretical framework is Q.E.D.

My attention returned at that time to the Pd/H and Pd/D systems. I had realized since the end of 1947 that these were the most extraordinary examples of electrolyte solutions. At that time I had found the early papers of Coehn, who had already shown in 1929 that H was present as H^+ in Pd host lattices (deuterium had not been discovered at that time). Moreover, the H^+ species was highly mobile in the lattice and, as the mobility obeyed the Nernst-Einstein relation, the species had to be present as "bare protons." In point of fact, the system behaves as an extremely dense plasma of protons (concentrations ~ 100 M) present within an ever higher concentration of electrons ($\sim 1,000$ M).

The investigation of Coehn led to a number of very uncomfortable questions about the properties of the Pd/H and Pd/D systems. Evidently, the hydrogen ions have to be extremely strongly bound in the lattice so that the dissolution process can be exothermic (*i.e.* H^+ in the lattice is more strongly bound than in H_2). At the same time, the hydrogen ions are highly mobile, a conundrum which defied resolution within the framework of Q.M. (This is an example of the type of inconsistency referred to in Appendix A.) There was also the further question: would it be possible to change the potential energy of D^+ in the host lattice sufficiently (by means of applied electric fields) to induce nuclear reactions? My answer at that time was "no" (based on the available Q.M.) except possibly under "heroic conditions."

The matter rested there until the 1960's, when I came to realize that the Pd/H and Pd/D systems had to be modeled using Q.E.D. At that time, we started a number of haphazard investigations of the Pd/H system. The question of whether one could induce nuclear reactions became more clearly defined at the end of that decade. Work on the isotopic separation of H and D showed that it was necessary to assume that the H and D present had to be modeled as many-body systems in order to explain the macroscopic behavior. I assembled equipment to start work on the putative nuclear processes on two occasions, but each time decided that such research would be judged as being inconsistent with holding an Academic Appointment!

The matter rested there until 1983, at which time Stan Pons and I discussed a number of options for further research. We

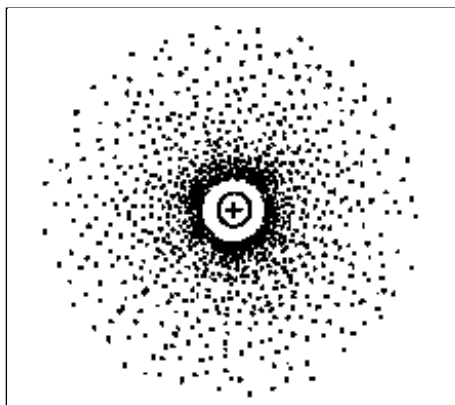


Figure 1. Schematic diagram of the negative space charge around a central positive ion formed by the electrostatic interactions of the ions.

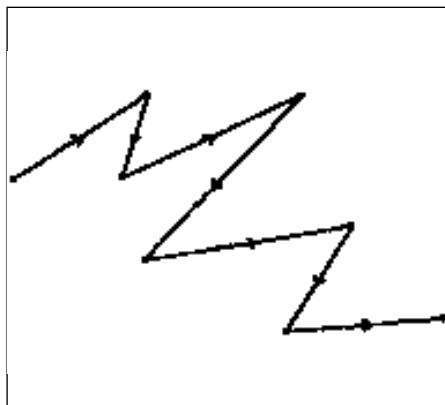


Figure 2. Schematic diagram of the Brownian movements of an ion. The arrows connect the points at which the ion is at rest.

believed that we could add one further major project to the topics then being investigated by our research groups. The topics we considered included:

- (a) relativistic effects in chemical reactions;
- (b) extension of the investigation of the structure and spectroscopy of interfaces;
- (c) the behavior of electrons in metals;
- (d) nuclear reactions of D^+ in metal host lattices.

Of these projects, we decided that (a) was beyond our means; and (b) was dependent on obtaining major funding which we could not secure. Of the remaining two projects, (c) was our first choice, but it rapidly became apparent that this, too, was beyond our means.

We therefore embarked on (d) and considered the implications of carrying out this project. As I have already noted, I had previously excluded research work on this topic; however, I had by that time resigned from my full-time academic position and, therefore, felt free to tackle (d). At the same time, the situation facing my colleague would clearly be serious if the nature of this project ever became known. We decided that the project not only had to have a "hidden agenda," it had to be totally hidden. This was all the more necessary because the military applications of any positive outcome of the research were not at all clear.

Start-up of the Project

As is well-known, we posed the following two questions at the outset:

- (1) Would the nuclear reactions of D^+ compressed into host lattices be different than the reactions in a dilute plasma (or than the reactions of highly excited D^+ in solids)?
- (2) Could such changes in the reactions be observed?

We expected the answer to (1) to be "yes;" thus, as the D^+ in Pd-host lattices is present in a quantum system of macroscopic dimensions, the rates of reaction would inevitably be enhanced. However, we expected this enhancement to be insufficient to allow the observation of (2), so the answer to this question was likely to be "no." Nevertheless, we started a limited investigation and considered experiments based on the options A, B, C and E:

- A: compression of D^+ in the lattices using electrodiffusion;
- B: compression of D^+ in the lattices using electrochemical charging; the effect of plasma excitation on such systems or on A coupled to B;
- C: charging of lattices by means of highly reducing/superbasic media;
- D: highly oxidizing/superbasic media and the link to "Hot Fusion" (see below);
- E: composite Systems, e.g. B or C linked to A.

Of these systems, A was our first choice, but we started with B (as a preliminary to A), because we believed that such systems are closest to the dictates of Q.E.D. (a view which was mistaken) and because electrochemical charging appeared to offer the easiest way of raising the potential energy of an extended Quantum System in an energy efficient way. Systems of type D were added to our list when it became clear that the nuclear reactions of D^+ in host lattices were radically different to those observed in "Hot Fusion" (see below).

As is also well-known, we opted for calorimetry as our primary "catch-all" methodology. Calorimetric methods can be made to be nearly as sensitive as the detection of nuclear parti-

cles and, indeed, are used in nuclear physics when it is necessary to make absolute measurements (e.g. in the estimation of Pu). Furthermore, the use of calorimetry was consistent with our wish to follow a "hidden agenda." The calorimetric method chosen had to meet a number of important criteria which included: conformation to "ideal behavior" (implying predictability of the performance from the Laws of Physics; we opted for the "well-stirred tank" familiar from the field of chemical engineering); high stability of the thermal impedances; uniformity of the temperature throughout the volumes of the cells, possibility of non-isothermal operation; high precision and accuracy and low unit cost—

we were financing the projects personally. These criteria dictated the choice of isoperibolic calorimetry using modified Dewar vessels.

The Initial Results: Sauce for the Goose and Sauce for the Gander. Serendipity, the Path to March 1989

The overall structure of the problem had become reasonably clear by the summer of 1988. We were observing the generation of heat in excess of the enthalpy input to the cells and far above that commensurate with the generation of tritium and neutrons predicted by measurements on "Hot Fusion." Moreover, the excess enthalpy was far beyond that which could be attributed to any parasitic chemical reactions. It appeared, therefore, that it was possible to establish nuclear reactions in quantum systems of macroscopic dimensions (following the dictates of the Q.E.D. paradigm), which not only had much higher cross-sections than those predicted on the basis of two-body processes, but which also differed in kind from those observed for two-body reactions. Evidently, it would be necessary to establish the nature of the reaction path(s). The detection of the most likely product, ^4He , in the presence of a huge excess of D^+ would be a project of the utmost difficulty and quantitative correlations of the yield of ^4He with excess heat production even more so. (We obtained our first indications of the formation of ^4He at later dates, but considered these results to be unpublishable.)

This lack of "nuclear ash" has proved one of the main points of criticism of the many "skeptics." However, our initial consideration of this question led to a further series of problems best described by the epithet "what is sauce for the goose is sauce for the gander." Thus, to the best of our knowledge, the very low yield of ^4He in "Hot Fusion" (about 10^{-6} that of the yields of neutrons and tritium) appears to be based on the low yields of energetic γ -rays associated with the formation of ^4He in the two-body processes. While such a conclusion may be valid for nuclear reactions in dilute plasmas, it may be far from valid for "Hot Fusion" in solid phases (more especially for "Warm Fusion" in such systems). In fact, the work on "Hot Fusion" must be judged to be incomplete in the absence of precise thermal balances and correlations of the yields of ^4He with that of the production of γ -rays. It was therefore evident that the project would need to be extended to include:

- D: Highly oxidizing/superacid media and the link to "Hot Fusion."

Our initial results (and the results obtained since 1988) appeared to indicate that there might be useful civilian applications without any military uses, a conclusion which ran counter to our initial expectations. However, closer reflection also showed that such a prediction might be incorrect. The system

One outcome of this research topic has been the demonstration that the majority of scientists are incapable of including serendipity in their thought processes.

we had developed was based on the premise of the Q.E.D. paradigm and it was not especially difficult to specify the changes which would need to be made to ensure that the system would converge onto the Q.M. paradigm — although we have never been questioned about this aspect and certainly have never had any intention of investigating the required changes.

In the summer of 1988 it had become clear that much further work was required, that the work would have to be broadened and that with an achievable acceleration of the program we might be able to assess the overall results by the autumn of 1990. We estimated that the cost of such an accelerated program would be ~ \$600K which was above that which we could meet personally. At that time we also believed that we had reached a stage at which it was necessary to inform the United States Department of Energy and the United Kingdom Atomic Energy Authority of the nature of our research. This would in any event have become necessary at some stage during the research, but we decided that it was opportune to combine the information with an application for funds for the envisaged accelerated program. At that time we made it clear that we were unable to judge whether the work should be classified, at least for a defined period of time. We believed that we ourselves did not have the necessary information to reach a decision on this issue, but that, in any event, it was necessary to continue a secret program until such time as it would be possible to carry out a complete evaluation of the project. However, it transpired that the restrictions which we wanted to impose ran counter to the funding policies of the Department of Energy.

It is relevant to consider here the role of serendipity in this research (indeed, in any research). It has frequently been asserted that these discoveries were made “by serendipity.” This view is incorrect although serendipity certainly played a part in the progress of the work. In my view, the true role of serendipity is the recognition of the significance of unusual results. It is better to guide one’s research by a series of logical steps rather than to indulge in a process best described as “Gee Whiz.” However, it is also important to accept and explain unusual results rather than to ascribe them to unspecified errors. One outcome of this research topic has been the demonstration that the majority of scientists are incapable of including serendipity in their thought processes.

The Position in March 1989

The outcome of our application to the Department of Energy can best be described as leading to the “worst case scenario” for our future research work: the whole matter was evidently going to be forced into the public domain. We therefore had to disclose our results to the administrative authorities of the University of Utah, who, in turn, felt bound to apply for Patent Protection. It is important to recall that we had by that time reached specific rates of energy production roughly equal to those in gas cooled fission reactors. In turn, the patent applications became the driving force for future actions.

The fact that this scenario followed this disastrous course has frequently been criticized. However, one must ask: “Can one imagine that the events could have followed any other scenario once the research was going to be driven into the public domain?”

I believe that it is important here to outline some of our reasons for wishing to delay the publication of the results. In the first place, although we had indications for the formation of ${}^4\text{He}$, these results were not publishable; secondly, we believed that most scientists would judge the work on the basis of the Q.M. paradigm and would therefore conclude that the results had to be false; thirdly, we did not believe that industry would con-

clude that research in this field (let alone any products based on this research!) would be in their short or medium term interest, although there might well be initial flashes of enthusiasm; fourthly, we believed that those concerned with national security could hardly be expected to welcome such research in the university sector; fifthly, we believed that we would lose our freedom of action because the research would become constrained by targets ill-matched to achieving further progress; lastly, we really wished to return to the more general problem of searching for examples of the operation of the Q.E.D. paradigm.

Post March 1989

As was to be expected, the topic of the nuclear reactions of D^+ in host lattices has turned out to be much more complicated and extensive than was apparent in March 1989. These are matters which will be described in other articles contributed to this issue of *Infinite Energy*. Furthermore, it has become apparent that the systems are “pseudo-simple”: while they are relatively simple to set up, their behavior is actually very complicated. Most of the key variables are “hidden” and are difficult to evaluate because the systems are only subject to very few controllable parameters (cell current or voltage, temperature for any given material).

As far as the generation of excess enthalpy using scenario B is concerned, it has become apparent that the production of low levels of excess enthalpy is easy to demonstrate in the early stages of the experiments provided the experiments are carried out with high precision and accuracy and provided satisfactory electrode materials are used. These provisos have turned out to be critically important, problems which have not always been understood. With increasing time and/or temperature, the systems pass through a region of “positive feedback” in the sense that increases in temperature lead to increases in the rates of excess enthalpy production. This “positive feedback” greatly complicates the investigation, another matter which has not been generally understood. “Positive feedback” leads to much higher levels in the rates of excess enthalpy generation, including the sustained production of heat at the boiling points at rates roughly equivalent to those achieved in fast breeder fission reactors. Furthermore, it has become apparent that it is possible to construct a number of systems operating above and beyond the onset of “positive feedback” and which generate excess enthalpy at lower levels but at zero enthalpy input. These phenomena have been described with a number of epithets including “Heat-after-Death,” “Heat-after-Life” and “After-Effects.” The work of Mengoli *et al.* on this topic is especially noteworthy.¹

The use of devices based on “Heat-after-Death,” etc. appears to open up the route to a range of “niche applications.” The work on systems of type B appears to open up the way to a much wider range of applications, especially to those which require the utilization of low-grade heat. However, it has been shown that much higher specific rates of excess enthalpy production (in the range $10 - 100 \text{ kW cm}^{-3}$) can be achieved using systems of type A (note especially the work of Preparata and Del Giudice).² Systems of this type may well lead to a very wide range of applications.

Appendix A. Choosing the Correct Paradigm: The Influence of Paradigms on Scientific Research

The example I have used in the main text (the use of electrostatics in the Debye-Hückel Theory) is a useful illustration of the influence of paradigms on scientific research. We understand the world in terms of models which are based in turn on

particular paradigms. I believe that all scientists would agree that, in constructing such models, we should use the simplest possible paradigm sufficient for the task at hand. Thus, the Debye-Hückel Theory leads to the interpretation of the variation of the self-energy of electrolyte solutions with the concentration. We do not need to use a more complicated paradigm as long as our attention is limited to the thermodynamics of such systems (albeit at very low concentrations).

At the same time, there are dangers in such a minimalist approach. The paradigm (and the associated models) come to dominate our thinking so that it becomes difficult (perhaps even impossible) to ask whether deviations from any predicted behavior may not be due to the use of an invalid paradigm? We do not ask the question: what would be the consequences of using a different paradigm? Instead, we attempt "to save the paradigm," with increasingly improbable special assumptions. Furthermore, the range of application of the paradigm is frequently extended to the interpretation of properties (or other properties of other systems) where its use is restrictive (possibly even incorrect). Thus, for the illustration used in this article, electrostatic models have been used in the interpretation of the dynamical properties of electrolyte solutions. Inevitably, such interpretations are kinematic: the consequences of using dynamic models remain hidden from view.

Evidently, we need some methodologies for choosing appropriate paradigms, a problem which has been much discussed in the philosophy, history and sociology of science. Such discussions usually center on the question: Were there (or are there) an increasing range of phenomena which could not (or cannot) be explained by an existing paradigm? We should also discuss the question: Can we arrive at "better interpretations" of given phenomena by changing the paradigm? However, this second question could open the way for much musing of doubtful value. It has, therefore, always seemed to me that we should use more "hard-nosed" approaches and I have found two such approaches to be especially useful.

In the first of these approaches, we can ask the questions: Can a given property A be interpreted in terms of paradigm X by using the argument (a)? Can a second property B be interpreted in terms of paradigm X by using the argument (b)? Such a list can be extended to include further properties. We then ask the further question: Are (a) and (b) self-consistent? If this is not the case, then the cause is likely to be the inapplicability of paradigm X. It is of interest that the dissolution of hydrogen and deuterium in palladium affords several examples of such inconsistencies if the paradigm X is Q.M.

In the second approach, we investigate whether the application of the paradigm leads to violations of the Second Law of Thermodynamics. I believe that most scientists would agree that the inability to violate this law (or else of some related principle such as microscopic reversibility) is our first line of defense against the development of invalid interpretations. However, it is also clear that whereas scientists are very adept at criticizing engineers, members of the public, or even each other (!) for the invention of devices based on Perpetual Motion, they are not very adept to subject paradigms and their derived models to the same scrutiny. The main text contains an illustration of this principle which led to the conclusion that processes in condensed matter must be interpreted in terms of Q.E.D.

Appendix B. The Search for Demonstrations and the Need to Invoke the Q.E.D. Paradigm

In the 1960's it appeared to me that it should be possible to devise experiments which would demonstrate the need to

model the behavior of condensed phases in terms of Q.E.D. rather than in terms of C.M. or Q.M. At that time (and at various times since then) I discussed some of the problems involved with other scientists whom I placed in two broad categories: those familiar with Q.E.D. and those, who, as far as I could judge, were not familiar with this particular field. Such a classification was indeed useful: the response of those in the first group to the problems raised by the modeling of electrolyte solutions was always immediate: "You must use Q.E.D."

The response of those in the second group was uniformly negative and, frequently, quite violently so. I came to believe that any research on this topic would have to follow a "hidden agenda." The topics had to be chosen so that they could be justified in terms of foreseeable interpretations in terms of C.M. or Q.M.; the specification of Q.E.D. at the outset would simply lead to sterile debate which would frustrate the research. The need to invoke this paradigm would, therefore, have to emerge from the interpretation of parts of the problem which were not essential to those parts leading to interpretations based on C.M. or Q.M., i.e. it was necessary to be able to say "given that the systems behave in the way seen, we can then use C.M. or Q.M. to reach the following conclusions." Q.E.D. would be restricted to the interpretation of the statement: "Given that the systems behave in the way seen."

The particular approach, which I favored at that time, was the study of the dynamics of very small systems as revealed by the direct observation of the fluctuations of the properties. (This will explain in part my interest in the study of electrodes having very small dimension, the so-called "microelectrode.") In the fullness of time we could specify four systems which had the potential of satisfying the "hidden agenda." Two of these systems were investigated but research on one of these had to be abandoned because of the pressure of work on "Cold Fusion," evaluation of the data sets for the other system was not completed for the self same reason (specifically, these aspects which required interpretation by Q.E.D.). In addition, there were nine further projects, of which four were investigated. However, the interpretation of the results for these systems were (or were expected to be) dependent on deviations from the predictions based on C.M. or Q.M. The demonstration of the applicability of the Q.E.D. paradigm was, therefore, not clear cut. It became evident that this line of investigation suffered from a major drawback in that the present day theory of stochastic processes is quasi-classical because the effects of Q.M. are only included to the extent of the recognition of distinct quantum states. Progress with the topics was, therefore, dependent on the development of the theory of stochastic processes based on Q.E.D.

Our attention was therefore directed to systems where the applicability of the Q.E.D. paradigm would become apparent from more macroscopic observations. (An approach which had actually preceded that based on the measurement of fluctuations in small systems and which led to the realization that electrolyte solutions have to be modeled using this paradigm. This aspect will not be described here.) The most extraordinary of these projects was based on the two questions:

- (1) Would the nuclear reactions of D^+ compressed into host lattices be different from the reactions in a dilute plasma (or from the reactions of highly excited D^+ in solids)?
- (2) Could such changes in the reactions be observed?

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