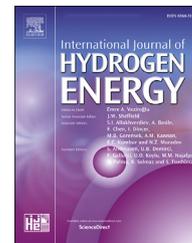




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Hydrogen and deuterium isotope effects beyond the electromagnetic force

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ABSTRACT

A mechanism is presented concerning electrolysis of deuteriated water with a palladium cathode that is infused with deuterium (palladium deuteride) resulting in the formation of small amounts of radioactive tritium, excess energy (more than allowed by EMF chemistry alone) and the concomitant liberation of ^4He . These net electron catalyzed nuclear chemical reactions ($^2\text{H} + ^2\text{H} + e^- \rightarrow ^4\text{He} + e^- + \text{heat}$) and ($^2\text{H} + ^1\text{H} + e^- \rightarrow ^3\text{H} + e^-$) appear to be a result of respectively four and three isotope effects acting in combination with each other in a non-linear (chaotic) fashion to produce a metastable nuclear isomer of hydrogen-4 or hydrogen-3. The four isotope effects begin with the influx of electrons into the $-\text{Pd}-\text{D}-\text{Pd}-\text{D}-$ Bravais lattice conduction band and consequent preferred rupture of individual weak Pd-D bonds (over those of Pd-H) in the cathode liberating D_2 . This is followed by the newly freed deuterium capturing an electron yielding a di-neutron (${}_0n_0n$). The ${}_0n_0n$ then reacts with a deuterium or hydrogen (from protic impurity in the lattice) via phonon enforced quantum tunneling resulting in ^4mH or ^3H respectively. The ^4mH quickly undergoes nuclear internal conversion to form ^4He . These reactions involve the weak force (Feynman Diagrams are shown), but they take place in simple electrochemical systems that are normally thought of in terms of the electromagnetic forces only. The combined influence of the four isotope effects explains thousands of, what were considered, anomalous observations by top electrochemical researchers. The newly described mechanistic effects involve a very important and almost forgotten intermediate (the di-neutron) and may even involve unique safety concerns.

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Introduction

Probably the quintessential pragmatic value of hydrogen would come from the development of the “The Hydrogen Economy”. The term was minted by John Bockris during a talk given at the General Motors Technical Center. The development of “The Hydrogen Economy” has been severely

attenuated by the relatively small amount of chemical energy per unit volume that hydrogen can deliver. ‘Bockris’ 1970 speech was delivered nearly two decades prior to his own realization that some unusual isotope effects involving our first element could possibly safely deliver orders of magnitude more energy than that via chemistry alone.

In these remarkable cases, a cursory understanding of quantum field effects governing nucleon–nucleon

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interactions is helpful. For example in the deuteron both nucleons are fermions, and a state containing two fermions must be anti-symmetric under exchange (obey Fermi–Dirac Statistics). The two nucleons (neutron and proton) are held together by the strong nuclear force. Since the strong force interaction between the proton and neutron does not distinguish between the charges, we neglect charge and consider spin. The deuteron (${}_0n_1p$) is a spin triplet, so a new symmetry is needed to obey the anti-symmetric necessity. Isospin is that symmetry, and ${}_0n_1p$ is an isospin singlet (see Fig. 1). Isospin was originally invented by Werner Heisenberg to distinguish between the proton and neutron.

The isospin symmetry is relevant to electrochemistry, because of the known possible capture of a relativistic electron by a proton in the condensed phase [1]. Similarly, the electrochemical capture of a relativistic electron by a deuteron would yield a di-neutron. The di-neutron is indeed a known observable species [2]. The electrochemical significance of this di-neutron has not been recognized, which is (in part) responsible for the disputation of a serious number of reports describing the electrolysis of deuterium embedded in a palladium cathode leading to the formation of a radioactive isotope (${}^3\text{H}$) [3–7]. These were solid empirical observations, and according to Storms [6] “Tritium appears to be the least ambiguous and most easily measured product” in this arena of electrochemistry. However, the consequential controversy raged in spite of the fact that these reports originated from major universities and national laboratories and are authored by important exemplary award winning electrochemical scientists. For example the Faraday Medal was awarded to John Bockris in 1981 and the Olin Palladium Award was bestowed upon Martin Fleischmann in 1986. The lack of an explanation for the tritium observations was the crux of the problem, but it was clearly of major value to these and other workers to report these products (e.g. ${}^3\text{H}$) prior to the construction of a viable mechanism [6,8]. The question is: how did the radioactive isotope of hydrogen get there? The question seems nonsensical in terms of the electromagnetic force only. The answer must begin with the formation of ${}_0n_0n$, which must be an isospin triplet and a spin singlet, as shown in Fig. 1.

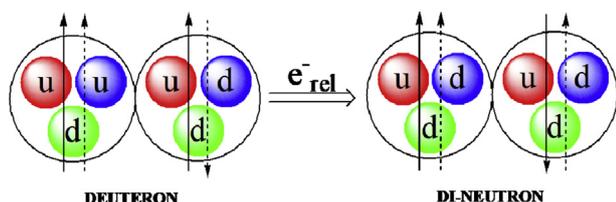


Fig. 1 – A simplistic representation of the two “spin” quantum numbers identifying the nucleons before and after capture of a relativistic electron. On the left, the deuteron is a spin triplet (both solid arrows up) and an isospin singlet (one dotted arrow up and the other down). Electron capture is symmetry breaking, and on the right the di-neutron is depicted as a spin singlet and an isospin triplet. Both ${}_0n_1p$ and ${}_0n_0n$ are anti-symmetric with respect to exchange. This is essential as ${}_0n$ and ${}_1p$ are identical with respect to the strong nuclear force. In the case of ${}_0n_0n$, spin does the symmetry breaking.

The electrochemistry of molecular systems containing deuterium is different than that of those of light hydrogen, for example the reduction potentials of deuterated molecular systems are less negative than those of their isotopically light analogues. The paramount historical example comes from the Urey and Washburn electrolysis of water to concentrate “heavy water” in H_2O . “Due to the easier electrolysis of water containing ${}^1\text{H}$ rather than ${}^2\text{H}$ or ${}^3\text{H}$, electrolysis can be used for producing water with reduced or enriched isotopic composition” [9,10]. Every bottle of purchased D_2O contains some tritium, and electrolysis enriches that isotope. However, this enrichment is quantitatively understood and accounted for, and the amount does not approach that measured in the Bockris, Fleischmann, Storms, etc. experiments.

Somewhat analogously, Goodnow and Kaifer [11] have shown that the electrolytic reduction of polyaromatic hydrocarbons occurs at more negative potentials for the perdeuterated analogues than for the perdeuterated systems. The Kaifer et al. work was in agreement with earlier solution phase chemical reductions where it was found that the added electron preferentially destabilizes the isotopically heavy molecule over that of the perdeuterated systems. In fact this isotope effect is just the beginning of a series of isotope effects leading to what is clearly the largest isotope effect in all of chemistry.

The overwhelming behemoth of all isotope effects is represented by the extraordinary observations made by Fleischmann [12] and Pons back in 1989 [13–16]. Martin Fleischmann was elected president of the International Society of Electrochemists in 1973. The year after he was bestowed the Olin Palladium Award, he garnered membership in the Fellowship of the Royal Society. The Fleischmann–Pons’ observations set off a fire-storm of acrimonious debate in both the chemistry and physics communities [17]. Using palladium metal cathodes that were pre-loaded with D_2 [7,17–20], they reported excess heat evolution during the electrolysis of deuterated water (D_2O) that was orders of magnitude greater than that obtained when using an ordinary H_2 – H_2O system. Often the electrolysis had to be in process for a period of days to months prior to the observations of excess heat. Fleischmann et al. did not reveal the preloading condition in 1989, which probably caused some of the initial reproduction attempts to fail [19]. In any event, the Fleischmann–Pons reports shook the chemistry and physics worlds to their cores, because more heat was reportedly released than possible via consideration of the electromagnetic force (normal chemistry) alone [21,22]. In spite of early reports of non-reproducibility [17,23,24], the Fleischmann and Pons Heat Effect (FPHE) has evolved into a contemporary and important reproducible phenomena [8,25,26], and a breakthrough theoretical understanding was initiated by Widom and Larson [1]. More recent experiments have rendered it increasingly clear that early failures [27–29] of reproduction were due to improper reenactment of the Fleischmann and Pons (FP) conditions, primarily attributed to improper pre-loading of the Pd cathode with deuterium [23,24,27–29]. A revealing detailed historical account is on record [28].

Indeed, achieving the necessary high loading of hydrogen and/or deuterium in the palladium has been a challenging requirement for the observation of chemically supra-thermodynamic FPHE. Pd can absorb vast amounts of H_2

and/or D₂ [30]. However in the early 90s, to obtain PdH ≥ 0.8, both elevated pressures and big blocks of time (from a day or two up to months) were required. More recently, electroplating the Pd on to a Pt electrode from PdCl in D₂O containing NaOD was shown to provide sufficient loading of D (or H) yielding prompt results [31]. After full understanding of the loading requirement, either pre-loading or electro-deposition, the FP experiment and consequent FPHE has been reproduced a plethora of times, many of which have been re-documented and cited in a recent book by Storms [8]. To quote Storms: “25 years of persistent study has resulted in overwhelming proof ... (page ix)” [8], and it is counterproductive to attempt to meticulously search for fault in each of the FPHE reproductions. It makes much more sense to provide a viable step by step mechanistic account for these empirical results involving nuclear physics. The over load of unexplained experimental data exists in the literature [7,8,13–16,18–22,24–29] and the abundance of empirical results needs to be unified prior to garnering more new data.

Electrochemistry, from its very beginning, has remained exclusively a study of phenomena involving the electromagnetic force of nature. Consequently, Fleischmann and Pons thought the ostensibly thermodynamically impossible excess heat must be nuclear in origin. However, the energy necessary to overcome the electromagnetic repulsion involved in forcing protons and/or deuterium nuclei sufficiently close together (< 2 fm) to allow the strong nuclear force to takeover would require hot plasma conditions. Further, the necessary fast neutrons released from fusion were not detected. The FPHE is actually an example of what is now called a Low Energy Nuclear Reaction (LENR) [8,26,32]. The very large (H/D) isotope effects involved in the LENR experiments are possible due to the low energy conditions. It has become apparent from a vast number of experiments that the excess heat effect is larger by more than an order of magnitude in the PdD system as opposed to the PdH system. In the latter system, the FP excess heat effect is usually not even detectable [33,34].

In FP electrolysis, electrons are galvanically added to a –Pd–D–Pd–D– lattice, and chemical isotope effects are fully anticipated. Indeed, electron addition to systems consisting of X–H and X–D repeating units, such as the X = C and presumably X = Pd systems, cause bond destabilization. Further, there is more destabilization in the isotopically heavy systems. Within the limits of the validity of the Born–Oppenheimer approximation, isotopic effects upon bond harmonic vibrational frequencies are simply related to the reduced masses of the protiated and deuterated systems [35–38]. The bond weakening isotope effects, caused by electron addition, result in the preferred Pd–D bond breakage over that of the Pd–H bond. Such Pd–D preferred bond breakage is the first of a series of isotope effects that eventually lead to involvement of the weak force as implied by Miles et al. and explained by Niebauer [39,40].

One of us, along with co-workers, have observed a very large solution phase chemical isotope effect upon the free energy (ΔG°) controlling the one electron transfer from the perprotiated hydrocarbon anion radicals to the correprotiated perdeuterated systems to form its radical anion [35–38]. Intrinsically, the added electron goes into an anti-bonding orbital which destabilizes the C–H(D) bonds.

Further, zero point energy effects necessitate that the added electron destabilizes the C–D bond to a greater extent than it does the C–H bond [37,41,42]. As a result, at 173 K, the one electron reduction of a mixture of benzene/benzene-*d*₆ was found to favor the formation of C₆H₆^{•-} over C₆D₆^{•-} by some 2.0 kJ/mol; the electron preferentially destabilizes the isotopically heavy molecule, hence $\Delta G^\circ > 0$ ($K_{eq} = 0.26$ at 173 K) [41] for reaction (1).



Further, this isotope equilibrium effect proves to be enthalpic in nature ($\Delta G^\circ = \Delta H^\circ = -RT \ln K_{eq}$ and $\Delta S^\circ = 0$) [35], and it is explained in terms of zero point energies as depicted in eq. (2) [41].

$$K_{eq} = \frac{\left(\prod_{i=1}^{j-1} e^{-\epsilon_i/2kT} \right)_{X_m D_n} \left(\prod_{i=1}^{j-1} e^{-\epsilon_i/2kT} \right)_{X_m H_n}}{\left(\prod_{i=1}^{j-1} e^{-\epsilon_i/2kT} \right)_{X_m H_n} \left(\prod_{i=1}^{j-1} e^{-\epsilon_i/2kT} \right)_{X_m D_n}} \quad (2)$$

In the case of the benzene system, $j = (3 \times 12) - 6 = 30$ vibrational degrees of freedom. The absence of an entropic contribution means that the bulk of the isotope effect is due to the weakening of the C–D bonds relative to those for the C–H bonds upon electron addition. These examples, and the Eigen values in eq. (2), refer to molecular systems. However, there are extensive similarities of the bonding in both the solid and molecular arena [43,44], a topic of significance here.

In the absence of a complete analysis of the mechanism of conductivity, bonding in the solid state is nothing particularly different from that in simple molecular systems; only the language is different [43]. In his review, Roald Hoffmann also wrote [44]: “In general, the interactions on a surface resemble those in molecules.” He further states: “shifts of electron density around the Fermi level, have bonding consequences.” In terms of the present discussion, this means that the electron count has an important bonding influence on the solid (cathodic) material surface that is analogous to what we have observed for molecules in solution.

The addition of electrons to the PdH and PdD systems requires that the added electrons go into anti-bonding orbitals (solution) or bands (solid, see Fig. 2). Electron addition destabilizes the individual Pd–H or Pd–D bonds, with the isotopically heavy systems (blue dots = D) destabilized preferentially. Solid state crystals of polyaromatic hydrocarbons, and their electron rich anion radicals serve as explicit examples of this added electron enforced destabilization [45–48]; the electron preferentially destabilizes the isotopically heavy band. The solution phase bond weakening upon electron addition carries over to the condensed phase as evidenced by the fact that naphthalene can be heated to 1200 °C without decomposition. However, warming of its anion radical crystalline salt, where the added electrons are now in a conduction band of the solid material, to just 100 °C results in bond rupture and hydrogen evolution [45–48]. When nitro groups are involved, electron addition decreases bond stability to the point of rendering shelf stable solids to very touch sensitive explosive crystals [46,47]; added electrons attenuate the barrier to bond rupture.

The point is that major bond disruption is realized upon electron addition and bonds involving D are more susceptible

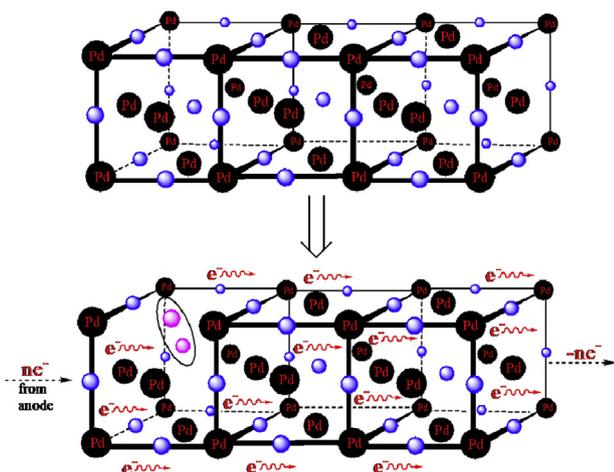


Fig. 2 – (Upper) A representation of three surface cells of the PdD lattice metal (large spheres). Note that the left-most cell has a face centered Pd atom missing. **(Lower)** Electrolysis is initiated changing the electron count, and Pd-D bond breakage takes place. In the region of the vacancy, the electron density is temporarily small enough to allow D₂ (small spheres) formation. The NAE develops.

than are bonds involving H [45–48]. The condensed matter Pd–H and Pd–D systems are not exceptions. The enhanced bond destabilization (*bd*) in the individual Pd–D bonds over that of the Pd–H bonds represents the first isotope effect (*IEbd*) ultimately leading to FPHE. *IEbd* accounts for the destabilization of six C–D bonds over six C–H bonds by 2.0 kJ/mol each, via the addition of one electron per 12 atom molecule. The effect will be much larger in the –PdD–PdH– system where a multitude of electrons are continually added across a conducting band. The added electrons are necessarily in anti-bonding bands, and since the relative bonding decrease is due to zero point energy differences (D relative to H), eq. (2) is applicable to X = Pd bonding systems, and the multitude of added electrons (from the external anode) weakens bonds in the –Pd–D–Pd–D– to a greater degree than in the corresponding –Pd–H–Pd–H– system. Empirical quantitative evaluation by carrying out isotope effect experiments with the Pd–H(D) systems, analogous to those carried out with the C–H(D) systems would be valuable. However, it isn't clear how such experiments could be carried out with a large solid lattice connected to an external anode and cathode.

In a large number of very carefully and elegantly engineered reproductions of the FP electrolysis (and alternative stimulation) experiments, LENR gives rise to excess heat, which has been unambiguously observed [8,25,26,33,34]. The number of reports is numerous, and they arise from very reliable and competent workers, but tedious details must be followed. In addition to the onerous amount of time for loading of the D₂ gas into Pd and the up to three month initial electrolysis that has hindered early workers, reproducibility has also been somewhat limited because of the difficulty in duplicating the (still unknown) effective sub-nano morphology and dimensions of the surface areas located at the nuclear active environment (NAE) where LENR takes place.

These problems have realized some amelioration from successful LENR observations using stimulations other than FP electrolysis [26,49,50].

Just four years ago, Erwin Lalik et al. [26] observed an oscillatory heat evolution reaction between O₂ and H₂ in the presence of Pd in a PAR (passive autocatalytic recombiner) type configuration. Further, on several occasions they observed a heat evolution that is nearly three times as great as is thermodynamically possible from the chemical reaction (2H₂ + O₂ → 2H₂O). As an explanation, they refer to LENR and the original FP publications. It should be noted here that the amount of excess energy realized by Lalik and other LENR workers is more than allowed by simple chemistry, but still orders of magnitude less than would be observed if a significant portion of the studied reaction were nuclear.

Discussion

In a typical FP LENR experiment, deuterium atoms are absorbed interstitially into the face centered cubic (FCC) Pd metal structure. Upon reaching PdD > 0.7, the β-phase becomes favored, and the overall density of the lattice decreases, concomitant with the mobility of D (or H). If the cathode is preloaded with D₂, the electrolysis generates multiple Pd–D moieties with extra electron(s) and consequent bond breakages leading to transient free deuterium atoms that are surrounded by electrons continuously being fed into the preloaded cathode via the external anode. Under these conditions, Hagelstein reasonably concludes that, following Pd–D bond disruption, “caged” D₂ molecules can form near monovacancies near the surface of the crystal lattice [51,52]. This is depicted in the left-most cell of Fig. 2, where the top face centered Pd is missing. It is thought that these regions form tiny spaces or nuclear active environments (appropriately termed NAEs) on the surface of the material [8]. The NAE cells containing these caged deuterons or protons are in a strong electric field during electrolysis, Fig. 2, thus allowing for the formation of D₂ (or H₂). Here, the interruptions of the Pd–D weak mobile bonds are enhanced over those of the Pd–H weak mobile bonds due to the aforementioned isotope effect (*IEbd*). Bonds to ²H are shorter than are those to ¹H [42]. This alters the general morphology of the respective condensed phase Pd lattice system. The very sensitive geometry of the NAE is thus perturbed via the presence of even a little ¹H, poisoning the LENR effect.

The formation of D₂ relies upon the release of two deuteriums in the same surface defect (cell) of the Bravais lattice. Until that happens, the steps leading to a LENR cannot begin. We could consider isolated Pd–D–Pd moieties as an unforced quantum harmonic oscillator with the D oscillating on the two frictionless springs (bonds) and obeying the Schrödinger eq. (3).

$$\left(\frac{-\hbar^2}{2\mu}\right)\left(\frac{\partial^2}{\partial x^2}\right)\psi_{xt} + \left(\frac{k}{2}\right)x^2\psi_{xt} = i\hbar\left(\frac{\partial}{\partial t}\right)\psi_{xt} \quad (3)$$

The corresponding time independent Eigen values are used in eq. (2). Our present case is clearly not that simple, because the oscillations are both dampened and driven by strong perturbations coming from three sources: 1. The bond

weakening in-flowing electrons; 2. Perturbing forces from the neighboring Pd–D–Pd vibrating strands; and 3. Phonons [53], one of the corner-stones of condensed matter physics. The other corner stone is electrons [53]. The differential equations for perturbed quantum oscillators are necessarily non-linear, because the time dependent perturbing differentials need to be added to the potential energy term in the Hamiltonian, which is only $0.5 kx^2$ in the unperturbed oscillator, eq. (2) [54].

After formation of D_2 , the intact D_2 molecule can re-associate with electron carrying palladium atoms. Nuclear electron capture by the (electron encrusted) caged D_2 of one of its orbital electrons would lead to a di-neutron and a single deuterium, as depicted in Fig. 3. Absent the Coulombic repulsion, the di-neutron can then undergo a nuclear reaction with this neighboring deuterium atom. However, the arithmetic reveals that a mass-energy problem exists, which is equivalent to about 780 keV. Ostensibly, the very high energies necessary for deuterium-electron capture are not present. Without this energy, the incoming electron cannot get closer to the nucleus than the Bohr radius, rendering the electron capture reaction (Fig. 3) exceedingly unlikely.

A free neutron has a finite half life ($t_{1/2} \sim 720$ s) before it decays into a proton, described in the well known Feynman Diagram shown in Fig. 4. As the down quark changes flavor, it emits a (weak force) W^- boson, which decays into an electron and an $\bar{\nu}_e$ antineutrino. In this case, the masses of the proton ($938.2720813(58)$ MeV/ c^2) and electron ($0.5109989461(31)$ MeV/ c^2) are 780 keV less than that of the neutron ($939.5654133(58)$ MeV/ c^2), and the excess energy is carried away by the antineutrino (see Fig. 4). On the other hand, a free proton cannot spontaneously decay into a neutron, because the neutron is “heavier” (has more energy) than a proton.

To allow the process to proceed to a FPHE, we need to overcome the arithmetic problem and garner sufficient mass-energy for an incoming electron, with the proper parameters, to be captured allowing neutron formation. This has been a

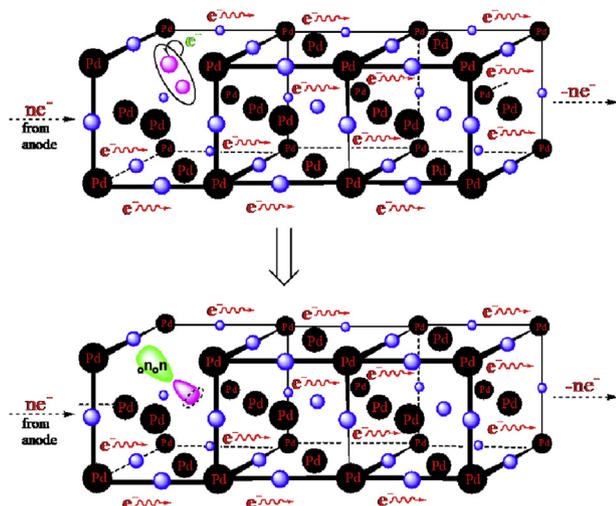


Fig. 3 – (Upper) A representation of the electron capture of a deuterium in D_2 , caged in a PdD lattice afforded by the vacancy. (Lower) This yields a di-neutron (left ovoid) and deuterium (right ovoid) with a single electron in close proximity.

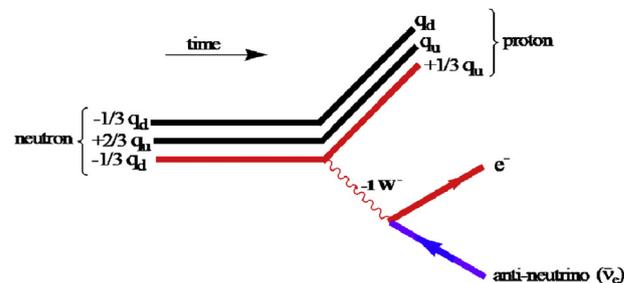


Fig. 4 – The Feynman Diagram for the decay of a down quark (confined in a neutron) into an up quark, electron, and an electron antineutrino. A W^- boson is created initially; it decays into an electron and electron antineutrino.

major stumbling block until Widom and Larsen [1] revealed that the electron mass in condensed matter can be augmented by local electromagnetic field fluctuations due to multi-pole moments. These fluctuations induce rapid fluctuating movement in directions other than that of the x translation of the electron though the lattice (electrochemical surface plasmons) [55]. Their treatment is classical, but it allows adding energy to the electron resulting in “heavy” electrons. Even if Widom–Larsen theory eventually fails to prove to be rigorously compatible with a rigid quantum mechanical test, quantum mechanics has already been shown to be compatible with relativistic electron formation in analogous mechanisms [56,57]. In essence, additional vectors of motion (y and z oscillations) imparts additional energy and enhanced relativistic nature to some electrons. The relativistic electrons, flowing through the lattice and interacting with the anti-bonding conduction band (in this case containing σ^* orbitals), can readily exchange with the bonding electrons of a D_2 molecule that is contained within the NAE. A simple calculation: $E = m_0 c^2 / \left(1 - \frac{v^2}{c^2}\right)^{0.5}$ reveals that the entire 780 keV arithmetic deficit is made up with an electron speed (v) at $0.92c$.

Relativistic electrons are known to cause otherwise unanticipated behavior in heavier elements such as the gold color of Cs and of Au and the liquid ambient nature of Hg. The smaller orbital occupied by the 6s electron is unavailable for bonding to nearby Hg atoms [58,59]. Widom and Larson treat the effects of the local field fluctuations in terms of the classical Hamilton–Jacobi equation, providing some mathematical rigor to the “heavy” electron capture and the formation of low momentum neutrons (Fig. 5). The neutrons formed in Fig. 5 have a very long DeBroglie wavelength, and consequently also have a large capture cross-section [1,56,57]. The electron actually captured (illustrated in Fig. 3) is a bonding σ electron, but recent experiments suggest that exchange between these and the σ^* “heavy” electrons can take place on the attosecond time scale [60,61].

When the blue spheres in Fig. 3 represent protons, as opposed to deuterons, one would anticipate a lower yield of low momentum neutrons simply due to the smaller electron capture cross section based upon size; that is an isotope effect. The charge radius of the deuteron is 2.1413(25) fm while that

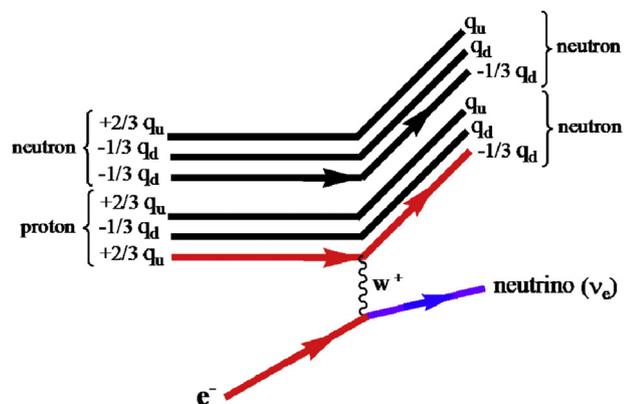


Fig. 5 – A Feynman Diagram for the capture of a relativistic electron (shown in bold) by a proton (two up quarks and one down quark) and decaying to a neutron (two down quarks and one up quark) and an electron neutrino. The breaking of isospin symmetry allows the newly formed neutron to become one member of a new di-neutron (${}_{\circ}n_0n$) of triplet isospin.

of a proton is only about 0.86 fm [59]. The electron capture (ec) by either nucleus is possible due to the weak interaction, as depicted in Fig. 5, and the respective difference in rate represents $IEec$ contributing to FPHE. Even though FPHE is more than an order of magnitude smaller in the case of the PdH system than in the PdD system [52,62], low concentrations of ${}^3\text{H}$ have been detected during the electrolysis experiments [3–7]. This is clearly due to protic impurity in the D_2O -PdD system [62]. The capture of a relativistic electron by D_2 yields ${}_{\circ}n_0n$, and phonon coupling leads to quantum tunneling combining the di-neutron with a nearby proton, if present, forming ${}^3\text{H}$.

Consistent with the proposed formation of transient ${}_{\circ}n_0n$, di-neutrons have been shown to actually exist for very short times outside of the nucleus as reported in Physical Review Letters [63]. However, they have been indirectly observed to live longer inside neutron-rich nuclei [64]. In the free state, the

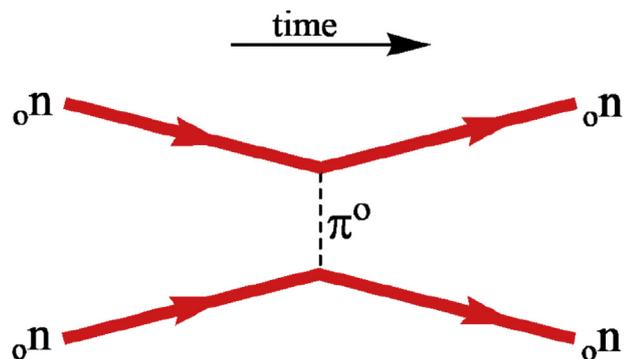


Fig. 6 – A Feynman diagram for the weak neutron–neutron interaction in the di-neutron (${}_{\circ}n_0n$) mediated by a neutral π -meson exchange. The individual quark constituents are not shown. Note that since these are low momentum neutrons with favorable isospin, the conservation of energy does not require their immediate scattering.

nucleon-nucleon interaction is mediated by the exchange of a neutral pion, see Fig. 6. The gyromagnetic ratio of an electron ($\gamma_e = 1.761 \times 10^{11}$ rad/sT) is orders of magnitude greater than that of a proton ($\gamma_p = 2.675 \times 10^8$ rad/sT). Hence, the electron capture and the consequent spin-spin scrambling (symmetry breaking) of the nuclear spin and isospin leave the majority of the resulting di-neutrons in the ground state spin singlet and the isospin triplet state [2].

The π^0 exchange, shown in Fig. 6, is different than is that due to W^+ , W^- , and/or Z boson weak force mediation [66]. The π^0 bosonic exchange has a range of only about 1 fm, theoretically rendering di-neutrons of border line stability in terms of the Heisenberg uncertainty principle. In the intimate proximity of the deuteron, longer residence times may be anticipated, which somewhat circumvent the Heisenberg stability restriction. The formation of the di-neutron results in a pair of two particle systems (${}_{\circ}n_0n$ and deuterium) in a lattice confined NAE where there is a phonon flow, see Fig. 7.

The di-neutron and neighboring deuteron are sufficiently close together so that phonon coupling can lead to quantum tunneling through the strong force regime producing ${}^4\text{mH}$. Hot ${}^2\text{H}$ - ${}^2\text{H}$ fusion reactions are known to depend upon quantum tunneling to get through the Coulombic barrier [67], but given the absence of a Coulombic barrier, the contribution from tunneling is unclear. Tunneling is so important in nuclear and EMF chemistry (and physics) that it has recently been labeled “The Third Reactivity Paradigm” [68]. We have some experience with quantum tunneling where no coulomb barrier is present, but this is in the absence of nuclear effects [69]. For the case of the extremely slow approach of the di-neutron to ${}^2\text{H}$ or ${}^1\text{H}$ in the NAE, both particles have very large DeBroglie wave lengths.

The four nucleon approach results in a large capture cross section for the reaction: ${}^2\text{H} + {}_{\circ}n_0n \rightarrow {}^4\text{mH}$. Normally the ${}^1\text{H}$ neutron capture cross section is almost an order of magnitude

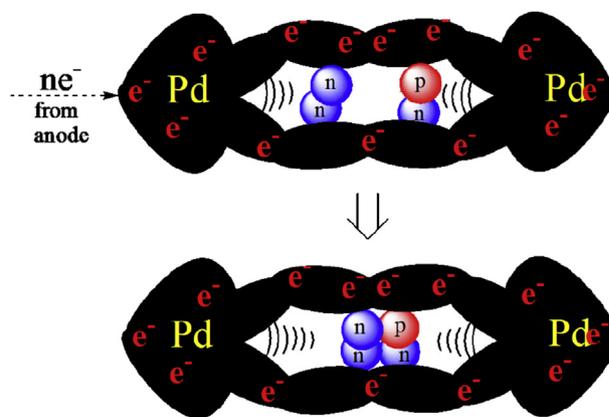


Fig. 7 – A cartoonish depiction of a deuteron and a low momentum di-neutron being phonemically compressed in a tight electron saturated NAE. It is anticipated that the nuclear isomer formed here has the di-neutron still intact, as described in Refs. [2,63,64], and a metastable nuclear isomer (${}^4\text{mH}$) is formed as opposed to ${}^4\text{H}$ created by ‘hot’ neutron bombardment of tritium, as described in Ref. [65]. Likewise, if the ${}_{\circ}n_1p$ were replaced by a single proton (1p), ${}^3\text{H}$ would be the product.

greater than that of ^2H , but this is primarily a result of the much greater scattering contribution from ^1H [70]. The scattering contribution is minimized under the very low particle momentums in the NAE. Consequently, the mutual capture (mc): $^1\text{H} + {}_0n_0n \rightarrow ^3\text{H}$ is probably slower than that leading to ^4mH ; this is IE mc . Di-neutron capture is not fully understood, but whether or not IE mc favors ${}_0n_0n$ capture by ^2H or ^1H is not of major importance here, because IE bd and the intentional loading of D_2 renders the concentration of $^2\text{H} \gg ^1\text{H}$. A very small neutron flux having been detected when a current was passed through a Pd electrode immersed in a deuteriated water salt solution [24] may serve as testimony to the fragility of the di-neutron. We argue that ^4mH does not yield a neutron flux but decays to ^4He : $^4m\text{H} \rightarrow ^4\text{He}$ [65].

The use of m to designate a metastable nuclear isotope is generally reserved for nuclei with half lives greater the 10^{-11} s or so, but the idea is analogous here. The point is that the hydrogen-4 nucleus that generates ^4He is not the same as that synthesized in the laboratory by bombarding tritium with fast-moving deuterium nuclei.

A common mode of decay of metastable nuclei (a nuclear isomer) is through *internal conversion* (ic) [58], where neutron to proton conversion and β^- ejection take place, see Fig. 4. After gentle (low energy) formation in the NAE environment, most of the ^4mH indeed undergoes rapid internal conversion yielding ^4He via beta decay. This is evidenced by the detection of ^4He concurrently with excess heat production in the FPHE experiment [32–34,52,62]. This is IE ic , Fig. 8. Free ^3H analogously decays ($t_{1/2} = 12.3$ years) into an isotope of helium (^3He) via beta decay ($^3\text{H} \rightarrow ^3\text{He} + \beta^- + \bar{\nu}_e$). The ^4mH decay process is different from that of ^4H obtained via the neutron bombardment of tritium, Fig. 8. The ^4H nucleus has a half-life of about 139 ys (1.39×10^{-22} s) and decays via neutron emission to tritium ($^4\text{H} \rightarrow ^3\text{H} + n$) [65]. Note here that some tritium (^3H) has

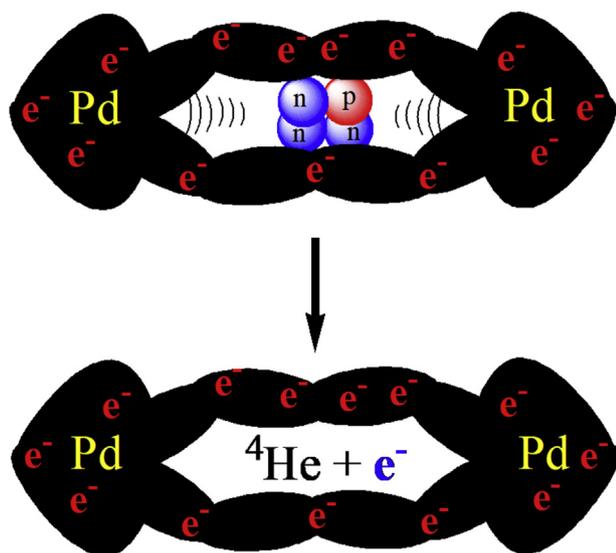


Fig. 8 – The newly formed ^4mH (top) subsequently undergoes β -decay into a ^4He plus an electron (bottom). The newly formed helium can garner electron(s) from the lattice to maintain neutrality. The actual volume of the reaction space is, of course, unknown.

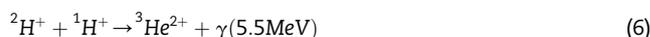
been detected in LENR experiments, but it probably arises from an electron catalyzed LENR between $^2\text{H} + ^1\text{H}$ (see Ref. [8] pages 13 and 225). Completely different modes of decay between the low energy nuclear isomer and the metastable isomer (as in the case of ^4H and ^4mH) are common. For example, in the case of the ^{58}Co and ^{58m}Co system, the former decays by electron capture and positron emission, and the latter undergoes gamma decay yielding ^{58}Co [58].

A critique of LENR was once discussed as “The Dead Graduate Student Problem.” FPHE experiments, producing just four Watts of excess heat and ^4He , should release more than 1018 neutrons per second and be fatal to the experimentalists [29]. The extreme lack of ionizing radiation is explained by the weak force decay of ^4mH as opposed to the strong force decay of ^4H . The strong force decay would be kinematically forbidden if the mass of the ^4mH , undergoing decay, is between that of a neutron plus a tritium and that of the ^4He nucleus (an α particle) plus an electron. This prescribed mass exists for the ^4mH decay in a rather narrow window between about 3.73 and 3.75 GeV/c^2 . Indeed, strong ionizing radiation is not observed and only feeble numbers of neutrons and alpha particles are predicted during FPHE experiments. Also, in this journal, recent well calibrated FPHE experiments revealed the un-questionable detection of a weak current of both of these entities [71,72].

McKubre et al. [32,73] found that the amount of ^4He ash is quantitatively consistent with the amount of ^2H consumed and heat generated, hence reaction (4) is thermodynamically balanced (IE bd + IE ec + IE mc + IE ic) and an electron catalyzed nuclear reaction. As described above, the tritium (^3H) detected in LENR experiments arises from an electron catalyzed reaction between $^2\text{H} + ^1\text{H}$, and IE ic is not involved.



It is also not the same nuclear reaction as found under solar or muon catalyzed conditions, both of which are fusion reactions. The analogous common hot fusion reactions that take place on the sun, reactions (5) and (6) involve the simple proton.



While the majority of the yield obtained from a fusion weapon comes from the fusion of ^2H and ^3H , the deuterium fusions in a thermonuclear weapon or a muon fusion are shown in reaction (7) and (8).



The hot fusion, not involving ^3H , found in thermonuclear weapons and muon catalyzed fusion have identical stoichiometries, (7) and (8). One would not expect the LENR reactions to be the same as those found in hot fusion, or even in “cold” muon catalyzed fusion, as LENR does not overcome Coulombic repulsion and does not involve hot fusion [74–76]. This unrealistic expectation was, however, utilized to criticize

[17,28] the now well recognized LENR observations reported by Miles et al. [40] McKubre et al. [32] and many others already cited [7,8,13–15,18,19,21,22,25,26,77]. FPHE and ^3H are probably best described to be a result of LENR that does not involve a hot fusion process.

Conclusion

To argue that electron(s) cannot approach closer to a hydrogen or deuterium in an NAE than allowed by the Schrödinger equation ($a_0 = 5.09 \text{ nm}$) is simply not a global constraint. Computer assisted humans cannot rigorously solve the Schrödinger equation (especially with relativistic contributions), even for well behaved heavy isolated atoms, such as Pd or Hg, let alone for nuclei cramped on the surface of a tight Bravais lattice NAE infused with a plethora of electrons.

As previously mentioned, subsequent to fully substantiating and reproducing the FPHE, it became apparent that other forms of stress could be applied to the ^2H or ^1H loaded Pd system to obtain LENR results. For example, simple LASER irradiation of PdD stimulates LENR [49]. Also, as already implied, the presence of oxygen (burning) also reveals an LENR response [26]. Experiments were carried out by simply heating palladium on carbon hydrogenation catalyst materials to 190°C under 3 atm of D_2 gas pressure [32,78,79]. The ^4He emissions were caught in leak-tight all metal vessel, eliminating possible contamination by ambient ^4He leaking through glass [78,79]. The ^4He arises, in the absence of a neutron flux, from the internal conversion of ^4mH most probably because: $m_{(4\text{mH})} < m_{(3\text{H}+\text{n})}$ otherwise ^4mH should (at least in part) decay to $^3\text{H} + \text{n}$ yielding a neutron flux, and $m_{(4\text{mH})} > m_{(\alpha+e^-)}$ as ^4He emissions are observed.

Excess heat energy and simultaneous ^4He evolution has been observed on a number of occasions. Gozzi et al. [80] presented bursts of ^4He evolution, in which bursts of excess energy from a Fleischmann-Pons type experiment were time-correlated with bursts that were passed into a gas stream for mass spectral analysis. Apparently any significant energy source added to the palladium deuteride (PdD) that is capable of stimulating a strong phonon response, and a corresponding phonon “gas” pressure in nano-crystals can result in LENR [81]. The phononic pressure [53] stimulates LENR and concomitant ^4He production as illustrated in Fig. 8. Reportedly, the LENR response can similarly, but with less reproducibility, be stimulated using metals other than Pd, for example Ti and Ni have been used [82].

It seems more than reasonable that the onset and profile of the LENR response is an example of a member of “the irregular, unpredictable behavior of deterministic, nonlinear dynamical systems” (a quote by Roderick V. Jensen) [83] playing out on the surface of the cathode. This is chaotic behavior analogous to the rhythmic dripping of a faucet suddenly becoming chaotic as described by James Gleick [84]. Information Theory reveals the impossibility of measuring unpredictability in going from the micro-scale to macro-scale due to extreme dependence upon the initial conditions of the micro-scale. In the case of FPHE, the morphology and condition of the NAE is an example of the mesoscopic scale, and the observation of LENR is manifested in the macro-scale. This is nothing new to

electrochemistry, or to quote Epstein and Showalter [85]: “Electrochemistry has been a very fruitful area for the study of chaotic dynamics.” LENR is a result of four isotope effects acting together in a non-linear fashion with an erratic phonon flow (produced by the stress of the incoming flood of electrons) to produce an example of a non-classical “Butterfly Effect,” which was made a household term by James Gleick.

In one of the Fleischmann et al. original articles, the temperature in their signature electrolysis cell, designed for the measurement of excess heat, remains close to 30°C for over $5.7 \times 10^6 \text{ s}$. At that point, it rapidly rises to 46°C then falls to 44°C then jumps to 60°C ; it then falls again and continues to oscillate up and down rapidly between 36°C and 47°C and does so continuously for over 5.5 days. It then falls again to about 30°C and remains there until the experiment is terminated several days later; see Fig. 8a in Ref. [7]. This ostensibly chaotic behavior is typical in a multitude of studies carried out by various (already cited) authors, and the measurements can be of heat, temperature, or ^4He release. Likewise photo emissions, emanating from the cathode, have been recorded during the electrolysis of $\text{PdCl}_2 + \text{LiCl}$ in D_2O [77,86]. These light flashes, observed in the IR region, are consistent with the electron capture as described in Fig. 3, and they are found to be randomly distributed in various locations on the palladium surface. Such observations further enforce the evidence that LENR is a surface phenomenon, and the macroscopic light flashes prove that a multitude of atomic phenomena nearly instantaneously take place in a finite area, and that the LENR event damages that area. Such destruction is evidenced by the appearance of “volcano” type pits in the surface of the anode [77].

There have been a number of unexpected explosions involving compressed mixtures of hydrogen and oxygen, where it is almost inconceivable that a spark could have been present [87,88]. A recent well publicized explosion, causing the loss of an arm, occurred where the researchers were carrying out biological investigations using accepted techniques [89]. The authorized explanation was an electrostatic spark. The gasses in these types of experiments are compressed into steel cylinders and pass through valves, which contain trace metallic substituents. Further, there are a number of examples where FP experiments result in mini-explosions where electrodes melt and equipment is damaged [13–15,77]. Could LENR have been a cause?

As stated above, palladium is one of the metals used in the PAR safety systems, the assembly of which is placed in nuclear reactor containment buildings. The assembly is exposed to hydrogen or deuterium for long periods of time. This raises a serious concern, for if a LENR spike were to occur due to the FP effect, an explosion could occur in close vicinity to the nuclear core. Indeed, this safety threat has been noted and experimentally investigated by Erwin Lalik et al. [26]. On several occasions they observed a heat evolution as high as the 700 kJ/mol H_2 . This is nearly three times as much heat as is thermodynamically possible from the chemical reaction.

It is difficult for us to imagine how such a large number of reputable and honored scientists (and some well known to fame) could all be mistaken in the empirical observation of the electrochemical production of atomic materials and heat that must have originated from reactions beyond the

electromagnetic force (currently called LENR) or even why such observations would appear to some to be heretical. Here we simply describe a reasonable mechanism involving well understood interactions (e.g. weak force interactions, nucleon electron capture, scientific chaos, and plasmonics) and normal particles (e.g. di-neutron and hydrogen-4) most of which are well documented in the literature. Further, the rather periodic, random, or even unreliable nature of the observables are simply a matter of scientific chaos. This mechanism explains the decay to and observation of ^4He , the commensurate (ostensibly supra-thermodynamic) heat production, the evolution of tritium, the observations of the weak neutron and alpha particle fluxes, and the annoying chaotic reproducibility, sustainability, and oscillatory nature of FPHE. Given the current interest in plasmonics [55], it is inevitable that the di-neutron and its effects will rise above the noise and reveal more new “supra-thermodynamic” measurements, and more otherwise unexplainable isotopes. Widom and Larson have given us some quantitative understanding of IEec. It will require major advances in condensed phase quantum field theory to get a mathematical understanding of IE_{mc} and IE_{ic}.

Finally, if LENR can be made to contribute to the futuristic “The Hydrogen Economy” it will have to gradually merge and convergence with other energy vectors [90]. Also, it is revealed by Weger et al. [91] that: “Such a transformation of our energy system will be a challenge, requiring new technological breakthroughs and renewable energy investments, and could take decades or generations to be carried out.” So it is, with the inclusion of pragmatic LENR, which would require massive scale up and advances in condensed matter quantum field.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ijhydene.2018.08.200>.

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