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Update on Neutron Transfer Reactions

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Abstract

We discuss progress in our studies of two new basic physical mechanisms that may be relevant to recent experiments that exhibit anomalies in metal hydrides and deuterides.

Anomalous energy transfer from phonon modes to the constituents of a lattice may occur through frequency-shifting phonon modes that are highly excited. The energy transfer is $\Delta E = N\hbar\delta\omega$, where N is the number of phonons in the modes, and $\delta\omega$ is the frequency shift of the phonon modes. A phonon laser can provide a large N; a finite frequency shift can be produced in a lattice with a phonon bandgap and with impurity vacancy modes that occur in the phonon bandgap. We propose that exothermic desorption in a metal hydride pumps a surface phonon laser.

We propose that neutrons can hop in a lattice, by analogy with electron hopping. Neutron hopping in crystalline silicon has been analyzed, and found to be unobservable; neutron hopping in lighter nuclei should lead to observable gamma emission. Neutron hopping in lighter nuclei, combined with anomalous energy transfer, is a candidate route to account for anomalous excess heat and tritium production.

1. Introduction

We have considered a number of possible theoretical approaches over the that past several years that may be relevant to a description of the anomalies that have been reportedly observed in metal hydrides and metal deuterides.[1] The primary experimental claims at this point include: [1] excess energy production in electrochemical experiments at a level that could not possibly be chemical (with essentially no associated radiation); [2] tritium production, without secondary neutron emission (the tritium, if real, must be born nearly stationary); and [3] neutron emission. There also exists claims of experimental evidence in support of

^{4}He production, host lattice activation, x-ray emission, gamma emission, and beta emission.

Many involved in the experimental efforts mentioned above hold that the existence of the anomlies is now proven; many involved in mainstream physics research (and most of the scientific community) hold that none of the anomalies are real. This difference of opinion appears to be a stable feature of the field, and there appear to be no prospects to resolve the differences any time soon. In our research, we take the experimental claims for which the evidence seems to be strongest, and study theoretical mechanisms that appear to be relevant. While the nature of the experimental claims excludes many possible mechanisms and reactions, they unfortunately do not generally make positive statements that would clearify reaction pathways or underlying mechanisms.

For any theory to begin to be relevant to claims of this sort, two fundamental issues must be addressed: [1] there must be a mechanism that allows for the transfer of a large amount of energy from the atomic scale to the nuclear scale; and [2] there must also be a mechanism that will allow nuclei to react in the most general sense. The second point is usually cast in terms of overcoming the Coulomb barrier, but in light of the discussion in the present work, this is too restrictive. Our studies have consequently focused on mechanisms for anomalous energy transfer, and also on new nuclear reaction pathways involving charge neutral reactants for which no Coulomb barrier problem occurs.

2. Anomalous Energy Transfer

We have considered during the past several years the possibility of anomalous energy transfer with the phonon modes in a lattice. Basic energy transfer mechanisms with vibrational modes in molecules and solids were understood in the 1930s; the creation and destruction of phonons in a lattice was described by Lamb[2] and later by Mossbauer[3]; frequency-shifting of phonon modes in the molecular case was described by Duschinsky[4]. Anomalous energy transfer through the creation and destruction of phonons does not appear to be feasible; instead, we have examined energy transfer by frequency shifting phonons that are already present.[5] In this case, the energy transfer is

$$\Delta E = N\hbar\delta\omega \tag{1}$$

where N is the number of phonons present, and where $\delta\omega$ is the associated frequency shift.

For this energy to be anomalously large, we require a large number of phonons N to be present (which could be developed by a phonon laser, for example). We also require a lattice with special properties: it must have a phonon spectrum that contains a band gap, and phonon modes must jump a band gap due to a modification of the lattice at a single site. Many metal hydrides contain a band gap between the acoustic and optical phonon modes; the basic theory for how phonon modes are altered due to mass or force constant changes at a single site was studied by Dawber and Elliott[6].

This mechanism alone appears to be capable of producing anomalies if the appropriate conditions can be met. For example, phonon gaps may be produced in metal hydrides if host lattice vacancies occur at impurity levels. In this case, processes that alter the number of host lattice vacancies can mediate anomalous lattice energy transfer. We have computed decay rates for this general process for a variety of recoil and nuclear decay channels.[7] For example, lattice-induced recoil of deuterons will lead to *dd*-fusion reactions with predicted neutron production rates that are in the general range of those claimed in experiments. This general mechanism could in principle produce anomalous alpha and beta decays; the associated reaction rates are reported in Ref. 7.

3. Proposed Surface Phonon Laser

Anomalous energy transfer through the route described in the last section presupposes the existence of a large number of phonons per mode N; we know that this could be produced by a phonon laser. Phonon lasers are known, and phonon lasing on acoustic modes up to 0.87 THz has been reported in the literature.[8] The energy exchange mechanism that we are interested in requires the existence of an optical phonon laser that operates near 6-8 THz.

Phonon lasers are in many ways analogous to optical lasers, as both require the development of a population inversion (or else the development of parametric gain). There exist few proposals for driving an optical phonon laser, although it would be reasonable to expect that optical pumping of impurities in a low temperature transparent crystal could be extended to optical phonons. Such techniques are not readily extended to metal hydrides, and the requirement that gain be developed at room temperature makes the problem far more severe since the optical phonon lifetimes are expected to be quite short (on the order of a picosecond) under these conditions.

We have recently proposed that exothermic desorption of a metal hydride can be used to drive a surface phonon laser.[9] If we consider the potential for molecular hydrogen near a clean metal surface,[10] we note that molecular hydrogen is not stable at the surface and is in fact pushed away (see Figure 1). Hydrogen in the metal can be either at lower or higher energy than the molecular energy. In the case that the energy is higher, so that the desorption is exothermic, then the situation becomes in some sense a phonon analog of an excimer laser.

Phonons can stimulate the desorption, and some of the desorption energy will go into the phonon modes that initially caused the desorption; in the event that the desorption is exothermic, then there exists the possibility of stimulated phonon emission and phonon gain. The hydrogen desorption from PdH probably becomes exothermic near a loading of 0.83; the situation is expected to be similar for desorption from PdD.

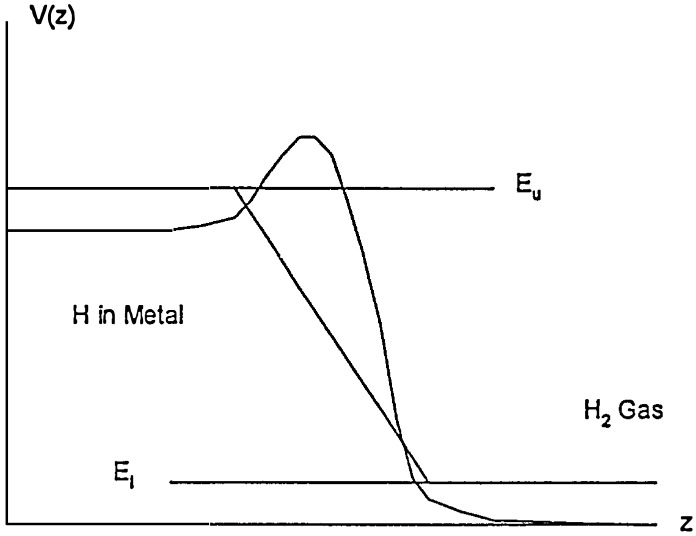

Figure 1. Schematic energy of H_2 minimum potential energy surface near a metal hydride surface.

Quantum desorption models have been studied in the literature (see for example Ref. 11). A basic description relevant to the present discussion could be developed starting from

$$\hat{H} = \sum_i \epsilon_i \hat{b}_i^\dagger \hat{b}_i + \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} \hat{c}_{\mathbf{k}}^\dagger \hat{c}_{\mathbf{k}} + \sum_{\sigma,\mathbf{q}} \hbar\omega_{\mathbf{q},\sigma} \hat{a}_{\mathbf{q},\sigma}^\dagger \hat{a}_{\mathbf{q},\sigma} + \sum_{i,j,\mathbf{k}} \left[V_{i,j,\mathbf{k}}(\hat{a},\hat{a}^\dagger) \hat{c}_{\mathbf{k}}^\dagger \hat{b}_i \hat{b}_j + h.c. \right] \quad (2)$$

This model includes hydrogen atoms at site i, free molecular hydrogen with momentum $\mathbf{k}$, phonon modes indexed by $\mathbf{q}$ and σ, and phonon-dependent desorption and adsorption terms. A linearization of the desorption potential in the phonon mode operators results in a Hamiltonian that can describe a one-phonon laser amplifier.

Perhaps the simplest quantitative estimate for the desorption flux required to sustain such a phonon laser comes from a balance between surface phonon creation and destruction; net gain is present when more phonons are generated than destroyed. If we imagine that a surface phonon mode is initially very highly excited, so that a significant fraction of the desorption power is converted to stimulated phonon power, then the threshold condition is equivalent to the statement that as many phonons are generated as destroyed. For surface optical phonons modes, the number of phonons required in this example is perhaps on the order of unity per atom participating; consequently the number of phonons destroyed per unit time is the product of the mode volume V, the relevant density n, and the destruction rate $1/\tau$. The number of phonons generated per unit time is the product of the exothermic desorption flux J_{des}, weighted by the number of phonons generated per desorption event $\overline{N}$, and mode surface area A. We obtain

$$\overline{N} J_{des} A = \frac{nV}{\tau} \tag{3}$$

The required desorption flux for normal surface optical phonon modes obtained from this argument is very high, on the order of 10^{28} molecules/cm^{2}sec. Such a desorption flux is not sustainable, even on a microscopic timescale, either by conventional diffusion or by thermodynamically-forced diffusion.[12]

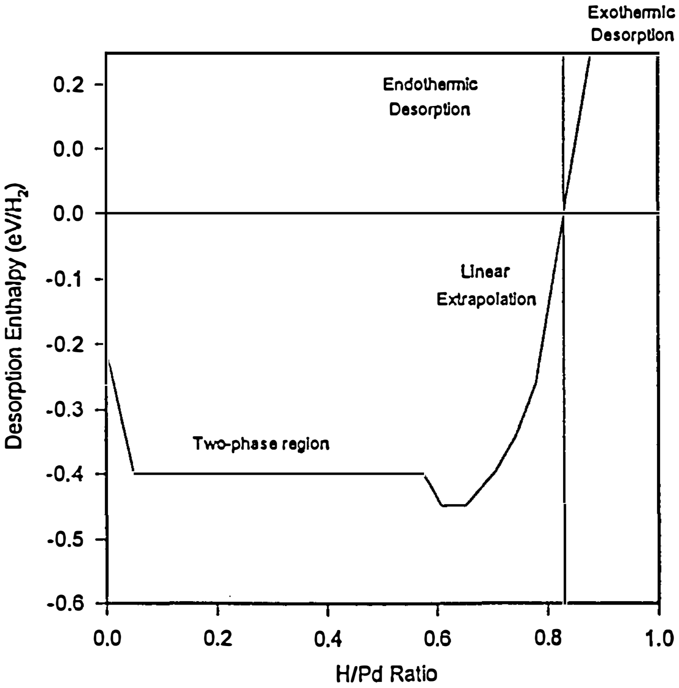

Figure 2. Estimated desorption energy for PdH versus loading.

Inspection of equation (3) indicates that the constraint on the desorption flux in the case of an impurity mode can in principle be lower by a dilution factor. For example, host lattice vacancies at the per cent level has been proposed to lead to an "impurity" optical phonon band within the PdD phonon band gap. The participating deuterium atoms are those within a cell containing a vacancy, which occurs at a reduced density; consequently, the threshold desorption current should be considerably less (on the order of 10^{26} molecules/cm^{2}sec). If a dominant mode of release of molecular deuterium from the surface of a highly loaded cathode is through local explosive exothermic desorption events that last on the order of a nanosecond, then such events may be sufficient to drive an optical phonon laser on dilute impurity modes.

The existence of such a mechanism would be critical for the operation of the physical mechanisms under discussion in section 2, and in the following sections. In addition this type of mechanism appears to be consistent with recent correlation of excess heat with flux discovered recently by the SRI group.[13]

4. Neutron Hopping

While there have been several proposals for mechanisms that may lead to enhanced screening of the Coulomb potential between nuclei, we have abandoned fusion reaction mechanisms generally (as candiates for heat and tritium production) in the belief that the amount of screening required is nonphysical. Our deliberations have driven us through a string of relatively exotic reactions, finally reaching a new class of reactions that may all be classed under the generally heading of "neutron hopping" reactions.[14] It is well known that electrons hop in solids, and the basic model that describes this hopping can be applied almost directly to examine the possibility of neutron hopping.

The neutron mixed valence Hamiltonian that describes the coupling between bound and continuum neutron states is

$$\hat{H} = \sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}} \hat{c}^{\dagger}_{\mathbf{k},\sigma} \hat{c}_{\mathbf{k},\sigma} + \sum_{i,\sigma} \epsilon_d \hat{d}^{\dagger}_{i,\sigma} \hat{d}_{i,\sigma} + \frac{1}{2} U \sum_{i,\sigma} \hat{n}_{i,\sigma} \hat{n}_{i,-\sigma} + \sum_{i,\mathbf{k},\sigma} \left[V_{\mathbf{k}} e^{-i\mathbf{k}\cdot\hat{\mathbf{R}}_i} \hat{c}^{\dagger}_{\mathbf{k},\sigma} \hat{d}_{i,\sigma} + V^*_{\mathbf{k}} e^{i\mathbf{k}\cdot\hat{\mathbf{R}}_i} \hat{d}^{\dagger}_{i,\sigma} \hat{c}_{\mathbf{k},\sigma} \right] \tag{4}$$

Neutron hopping is only expected in nuclei with *s*-wave valence neutrons and two neighboring stable isotopes; elements which satisfy this requirement include: hydrogen, helium, silicon, cadmium, tin, tellurium and xenon.

We recently investigated neutron hopping in silicon, in the hopes of developing an experiment that would exhibit the effect cleanly. The basic idea is that $2s$ neutrons from ^{29}Si would hop among neighboring ^{28}Si nuclei, and from time to time be captured by ^{29}Si to make ^{30}Si; this capture process would lead to a gamma at 2.1 MeV. Crystalline silicon has a diamond lattice structure, which consists of two interpenetrating FCC lattices. Virtual neutrons originating from one FCC sublattice are Bragg scattered by the other and delocalized; the gamma emission rate for this structure is obtained from fourth order perturbation theory

$$\Gamma = -\frac{2}{\hbar \epsilon_D^4} \mathrm{Im} \left\langle \hat{V}_{13} \hat{V}_{32} (E - \hat{H}_2)^{-1} \hat{V}_{23} \hat{V}_{34} (E - \hat{H}_4)^{-1} \hat{V}_{43} \hat{V}_{32} (E - \hat{H}_2)^{-1} \hat{V}_{23} \hat{V}_{31} \right\rangle \tag{5}$$

In this formula, state 1 refers to a ^{29}Si valence neutron on the first sublattice, state 2 refers to a ^{29}Si valence neutron on the second sublattice, state 3 refers to a free neutron, and state 4 refers to a gamma captured ^{30}Si neutron; the interaction potentials $\hat{V}_{ij}$ are of the form of given in the neutron mixed valence Hamiltonian.

This gamma emission rate can be evaluated approximately (after much algebra); we obtain in the high temperature limit

$$\frac{\Gamma}{^{29}\mathrm{Si}} \approx \frac{V_0^6 N_B^4 \gamma}{\epsilon_D^4 \delta\epsilon_M^2} f[^{28}\mathrm{Si}] f[^{29}\mathrm{Si}] e^{-\Delta\epsilon/kT} \tag{6}$$

where V_0 is the volume-averaged nuclear matrix element (about 20 meV), N_B is the number of Brillouin zones contributing (about 30 near the melting point), γ is the capture rate for a free virtual neutron (about 2.5 sec^{-1}), ϵ_D is the neutron binding energy (8.47 MeV), and $\delta\epsilon_M$ is the lattice energy transfer in the Mossbauer limit (uncertain, but perhaps on the order of 10^{-10} eV). The f factors are fractional site occupation probabilities. The exponential factor comes about from destructive interference of the continuum Bragg waves, limited by the requirement that the virtual neutron scattering generates no phonons; the effective energy barrier is

$$\Delta\epsilon \approx \frac{a^2 kT}{< |\mathrm{u}|^2 >} \longrightarrow \frac{2Ma^2(k\Theta_m)^2}{9\hbar^2} \approx 22 \text{ eV} \tag{7}$$

where a is the lattice constant (2.715 Å), $< |\mathrm{u}|^2 >$ is the mean square thermal center of mass displacement, M is the nuclear mass of ^{28}Si, and Θ_m is the Debye temperature (about 525 K). This relation may also be written as

$$\Delta\epsilon \approx \frac{1}{3} M\omega_0^2 a^2 \tag{8}$$

where ω_0 is the characteristic vibrational frequency.

The gamma emission rate in this case is unfortunately too small to be observable. The emission rate is highest near the melting point (1683 K); the prefactor is respectable ($\sim 10^{-13}$ sec^{-1}), but the exponential (due to destructive interference of the Bragg waves) reduces the rate by more than 60 orders of magnitude.

Consequently, we turn to the question as to whether the effect would be observable in other systems. Upon inspection of the various factors that make up the effective energy $\Delta\epsilon$ for the diamond lattice structure, it appears that only two quantities are of importance: the nuclear mass and Debye temperature. Lighter nuclei could exhibit in principle a much greater effect. Of the candidate elements with outer s-wave neutrons, by far the most interesting candidate is hydrogen: virtual neutron originating from deuterium have the best chance of being delocalized by Bragg scattering from nearby protons in an ordered lattice. This argument assumes that the relevant nuclear matrix element (which we have not yet computed for deuterium) is of comparable magnitude. For example, the vibrational frequency ω_0 for the deuterons in PdD is roughly the same as for the silicon nuclei in crystalline Si (the lattice structure is that of NaCl, so that a somewhat different proportionality may occur); the barrier energy $\Delta\epsilon$ in this case should be much less.

If these arguments are correct, then we would expect that a metal hydride that is loaded with a mixture of deuterium and hydrogen would exhibit neutron hopping if heated suddenly, as a thermally-induced bulk effect, with an effective energy on the order of an electron volt. The presence of the effect should be observable through gamma emission. Computations of this process are ongoing.

5. Neutron Hopping Coupled with Anomalous Energy Transfer

For the past several years we have examined various approaches to the problem of neutron hopping to an inequivalent nucleus coupled with the lattice energy transfer mechanism. A neutron hopping onto a nucleus generally will lower the phonon mode frequency, which leads to an energy transfer that has the wrong sign for heat production. We proposed that impurity modes due to host lattice vacancies could shift up through a recoil upon neutron capture that reduced the number of vacancies. While it seemed to be clear that the energy transfer through frequency shifting could be driven by a neutron transfer in principle, the frequency shifting mechanism leaves the lattice in a state which cannot use the same phonons for subsequent energy transfers.

This problem is in general serious. If phonons were used only once then lost, then the maximum efficiency would be much less than one; the experimental claims for heat production are larger than this. We have proposed earlier that the phonons could be frequency-shifted back down by a Raman process, and replaced when lost by phonon gain. While this may be possible in principle, it is by no means compelling.

Here we propose a speculative alternate solution that may have some advantages. We propose to couple an exothermic neutron hopping reaction with an endothermic lattice-induced decay process, and then require that the phonon distribution not be changed significantly during the combined process. Such a mechanism could be repeated without a loss of phonons; it has the additional advantage that the reaction rate for the coupled process will be faster than the rate for the neutron hopping part of the reaction alone, since the lattice-induced processes are so fast.

For example, we consider a virtual neutron that originates from a deuteron, resonantly Bragg scatters off of nearby protons in a metal hydride, and then lands on ^{28}Si with enough recoil to dislodge it so as to change the number of host lattice vacancies. If the process were conservative so far, then the lattice would be highly excited and unstable against a rapid decay of the host lattice, the fastest of which would be lattice-induced alpha decay of a low Z nucleus. Since the lattice decay is so fast, it is reasonable to couple it with the preceding reaction as a second order process,[7] which need not conserve energy in the intermediate state. The associated decay rate of a deuteron through this route can be computed from

$$\Gamma = -\frac{2}{\hbar\epsilon_D^4\Delta\epsilon_L^2}\mathrm{Im}\left\langle \hat{V}_{13}\hat{V}_{32}(E-\hat{H}_2)^{-1}\hat{V}_{23}\hat{V}_{34}\hat{V}_{45}(E-\hat{H}_5)^{-1}\hat{V}_{54}\hat{V}_{43}\hat{V}_{32}(E-\hat{H}_2)^{-1}\hat{V}_{23}\hat{V}_{31}\right\rangle \tag{9}$$

In this formula, $\Delta\epsilon_L$ is the energy excess determined by the difference in neutron binding energies of the donor and acceptor nuclei, taking into account the energy transfer with the lattice:

$$\Delta\epsilon_L = |\epsilon_A| - |\epsilon_D| - N\hbar\delta\omega \tag{10}$$

and state 5 includes the results of the lattice-induced decay.

This mechanism is currently the focus of our theoretical investigations. While we have not yet evaluated this rate in detail, certain aspects of the reaction rate appear at this point to be plausible. We expect the total reaction rate to be perhaps of the form

$$\frac{\Gamma}{D} \sim \frac{V_H^6 V_{Si}^2 N_B^4}{\epsilon_D^4 \Delta\epsilon_L^2 \delta\epsilon_M^2} f[\mathrm{D}] f[\mathrm{H}] f[{}^{28}\mathrm{Si}] e^{-\Delta\epsilon/kT} e^{-\theta} \sum_i \gamma_i \tag{11}$$

The prefactor here is reduced from that encountered in the last section by $V_{Si}^2/\Delta\epsilon_L^2$; the fractions f can be on the order of unity; the effective energy $\Delta\epsilon$ ought to be less than 1 eV. The probability that the appropriate recoils occur, and that the coupling between the initial, intermediate and final lattices is phonon conservative, will appear as an exponential damping factor (taken to be $e^{-\theta}$ here, the magnitude of which is presently unknown). Finally, the lattice-induced decay gives rise to a very large rate γ_i (on the order of 10^{20} sec^{-1} for each nucleus i in the lattice that is capable of causing a useful phonon mode gap jump; this results in a potential increase in the total rate over the results of the previous section.

One interesting feature of this type of theory is that it does not require that the lattice actually has the very large number of phonons required for a complete exchange of the reaction energy. Since the intermediate state is now virtual, and the final state lattice does not have the reaction energy (it is in the fast alpha in this case), the requirements on the lattice are now much reduced. Two requirements are seemingly apparent however: (1) given the combinatorial number of available phonon states, the lattice states with frequency-shifted phonon modes had best have large coupling matrix elements and be well separated in energy to avoid destructive interference effects; and (2) given the fast (psec) decoherence time of the optical phonon modes, unless phonon gain is present on the gap jumping modes, the number of atoms that can take place in the final alpha decay process will be limited. Further study of this mechanism will determine the applicability of the theory to experiments reporting heat and tritium production.

6. Conclusions

We have reviewed key aspects of our theoretical approach to account for anomalies claimed in recent experiments with metal hydrides. We have focused on two new physical effects: [1] lattice energy transfer by frequency shifts of phonon modes, and [2] neutron hopping. Neither of these mechanisms has been proven experimentally, yet we contend that both effects are predicted from quantum mechanics.

Specific results that are new since ICCF4 include: [1] a proposal for a surface phonon laser driven by exothermic desorption; [2] that neutron hopping should lead to second order gamma emission as a thermally-induced bulk effect; [3] the observation that Bragg scattering of virtual neutrons by the internal unit cell structure is important (especially for nuclei with outer *s*-wave neutrons); [4] the conclusion that the primary donor or acceptor nuclei for heat or tritium production must involve outer *s*-wave neutron orbitals; [5] that neutron hopping should work much better with light nuclei; [6] that the energy exchange with the lattice during a neutron hop in the Mossbauer limit is nontrivial and must be included properly. The formula for second order gamma emission, as well as the theory outlined in section 5 constitute new theoretical results.

We conclude that there exist routes within conventional theory both for anomalous energy transfer and for a new class of reactions based on neutron hopping. The theory outlined in section 5 describes excess heat and tritium production mechanisms, perhaps similar to the experimental claims; further calculations and comparisons with experiment are required to determine whether it is correct in detail. The phonon laser route to energy transfer outlined in sections 2 and 3 can result in neutron, alpha, beta and gamma production, also perhaps relevant to the experimental claims. The neutron hopping/gamma emission process outlined in section 4 is a bulk effect, and may be more easily verifiable experimentally.

Quantitative predictions specific to the various metal hydrides for thermally-induced second order gamma emission will soon be available; these predictions can be tested experimentally, and can be used to prove or disprove a major part of the present theory. Heat production according to the theory of section 5 a neutron transfer from deuterium to silicon, and a low *Z* alpha decay; this can be proven or disproven through measurements of silicon isotopic ratios, and detection of the alpha decay products.

Acknowledgements

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