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Research Article

# Microscopic Insights into the Anomalous Heat Effect that Unify Disparate Experimental Results

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**Abstract**

The anomalous heat effect (AHE) has many embodiments including hydrogen loading by electrolysis, high and low hydrogen pressure over elevated temperature nanostructures, ultrasound, and glow discharge loading. The AHE was triggered by variable charging current, temperature fluctuations, high voltage pulses, laser pulses, ultrasound, and electrolysis time. Pd materials for which the AHE has been reported include wires, cylinders, foils, and nanoparticles. Ni materials for which the AHE has been reported include constantan wires, Ni in zeolites, and Ni nanostructures. It is highly likely that the same AHE mechanism underlies all these embodiments despite their seemingly wide differences. Many investigators involved in research on the anomalous heat effect (AHE), including this author, are of the opinion that phonons in Pd and Ni play a role in generating and sustaining the AHE. Here it is hypothesized *that the AHE requires a specific frequency optical phonon resonance that couples to electromagnetic radiation of the same frequency. If it can be arranged to sustain this specific, resonance, then the AHE is produced.* It is demonstrated how this one assumption, coupled to several recent experimental results, can lead to useful microscopic insights that unify disparate experimental results under one umbrella and that may also be useful to guide further experiments.


*Keywords:* Anomalous heat, Phonons, Pd deuteride, Ni hydride

## 1. Introduction

It *is hypothesized that the AHE requires a **specific frequency** optical phonon resonance that couples to electromagnetic radiation of the same frequency. If it can be arranged to sustain this specific resonance, then the AHE is produced.* In this paper it is demonstrated how this one assumption can lead to useful microscopic insights that unify disparate experimental results under one umbrella. The ideas that led to this model came from perturbed angular correlation (PAC) results on palladium hydrides recently obtained by this author at CERN [1], recent x-ray diffraction data on superconducting palladium hydrides [2], and 40-year-old internal friction data on PdH [3].

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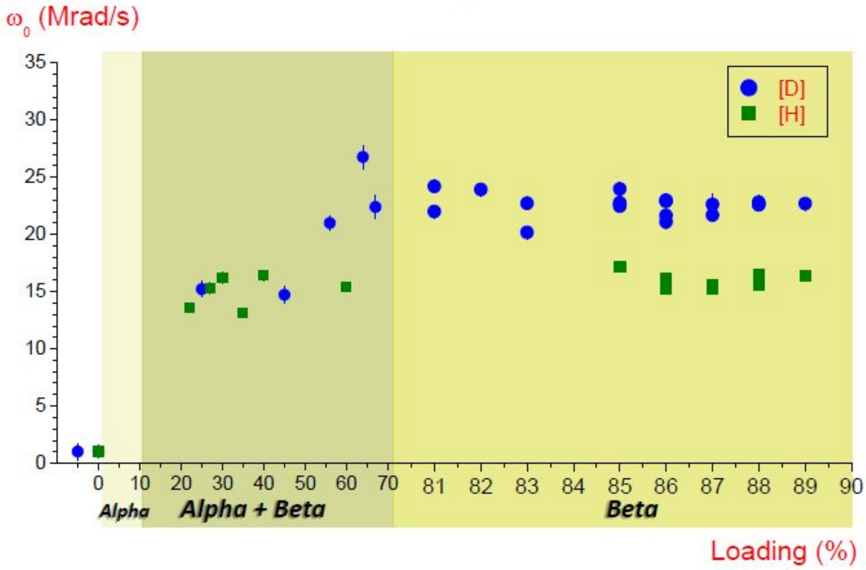


**Figure 1.** Precession frequency in Megaradians/s versus H or D loading fraction for dilute $^{181}$Ta in a Pd host lattice. Precession is caused by an electric field gradient at the Ta nucleus site [1].

## 2. Underpinnings of Model

A perturbed angular correlation (PAC) experiment was conducted at the ISOLDE facility at CERN where radioactive Hf ($^{181}$Hf, 42 day half-life decays to $^{181}$Ta + Beta, 11 ns half-life) was ion implanted into Pd foils and the gamma cascade decay of the daughter Ta nuclei were employed to measure, in situ and at room temperature, the local electric field gradient (EFG) around the nucleus as a function of the electrochemically loaded fraction of D or H [1]. A nucleus that possesses a quadrupole moment precesses in an EFG. The precession frequency of the nucleus due to the coupling of the nuclear excited state quadrupole moment with the EFG at the nucleus is a measure of the local strain around the atom. Figure 1 shows the precession frequency (strain) versus the loading fraction for both D and H. Note that the strain around the Ta atom that substitutionally replaced a Pd atom is 43% larger for D in Pd than H in Pd above ~0.5 concentration - a surprising result.

Syed et al. [2] prepared $PdD_x$ by soaking Pd in 100 bar pressure of deuterium at a temperature of 300 C, followed by quenching to 77 K and continued cooling to 40 K. A superconducting phase was found with a superconducting transition temperature, Tc, of 61 K (Fig. 2), another very surprising result since it has been known for 50 years that $PdD_{>0.9}$ has a Tc of 11 K. What's more, the nearly flat resistivity change with increasing temperature in Fig. 2 shows the phase was stable up to room temperature since the resistivity change for a normal metal increases sharply with temperature according to the Drude model. Since there was no concomitant crystal phase transition there must be an electronic phase transition to account for this result. X-ray diffraction determined that the quench froze in a ~30% tetrahedral site occupation of D that is not present in room temperature loaded Pd that has 100% octahedral site occupation. Noting that the tetrahedral position has 1/3 the volume of the octahedral position, Syed conjectured that a significant fraction of D occupation in the tetrahedral site would expand the spacing between Pd atoms which then alters the electronic band structure to provide the Tc of 61 K. The maximum T occupancy occurs where the flux of diffusing atoms is a maximum, all of which must pass through T sites. The authors state, "At high concentrations, the

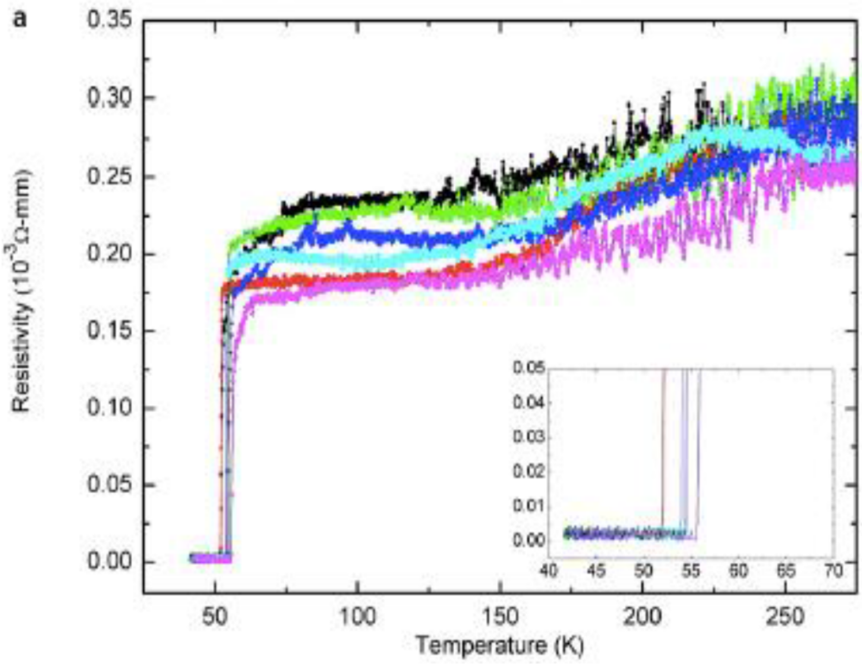


**Figure 2.** Resistivity versus temperature for 6 samples of Pd loaded with D at 300C and then quenched [2].

O sites are nearly filled, thus blocking the diffusion pathways. In the middle range, therefore, the diffusive flux and potential for T occupancy is greatest". For another example, the stretching of superconducting $MgB_2$ increases the Tc by a factor of two [3].

Figure 3 plots the internal friction data of Mazzola et al. [4] for $PdH_x$ from x = 0 to 0.89. Internal friction is a measurement of the mechanical coupling to lattice defects by noting the damping as a function of temperature of a resonator (1/Q, the resonator quality factor) made from the material of interest. The two internal friction peaks in Fig. 3 are for mechanical coupling to H diffusion and to dislocation motion. Note that the diffusion peak has a maximum at ~0.64 and then rapidly declines because diffusive jumps are blocked since the adjacent sites are occupied. A conclusion from this data is that if H diffusion dynamics are desirable for the AHE, then the loading should be less than ~0.64. To populate the tetrahedral site, diffusive jump dynamics are desired so, operating at reduced loading helps. Ed Storms has been saying for years that the AHE exists for loading as low as 0.3 [5]. Figure 3 supports this notion. A second conclusion is that dislocations are locked above about 0.55 loading.

Returning to the PAC experiment, the Hf was ion-implanted at 80 KeV energy so all Hf atoms are less than 20 nm from the surface where the D diffusion in and out of the surface and electron current dynamics are maximized. Therefore, it is reasonable to assume that electromigration could provide the energy necessary to elevate substantial D into the tetrahedral site so that the material synthesized by the quench of PdD from 300 C and the PdD in the surface created in the PAC experiment are *one and the same material*. Coming full circle, the larger strain measured in the PAC experiment is explained by surmising that ~30% tetrahedral occupation of D expands the Pd interatomic spacing which produces the measured strain. The larger zero-point motion of the hydrogen makes it less stable in the tetrahedral site than deuterium in the dynamic surface region so only deuterium shows the increase in strain.

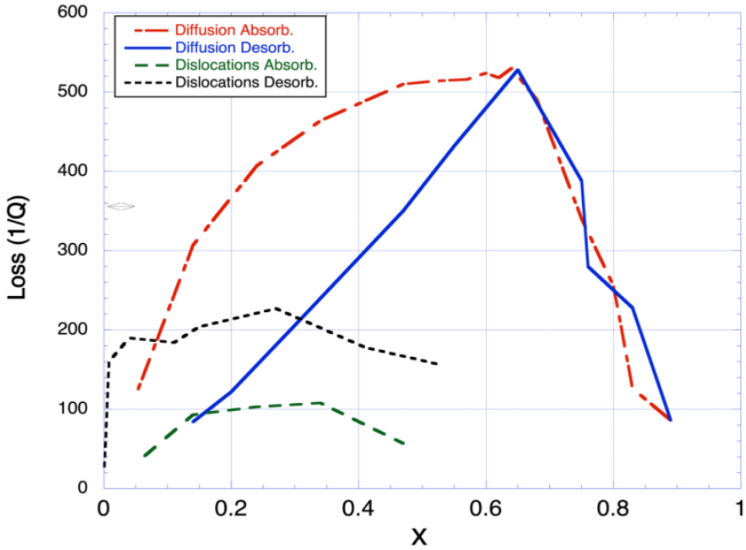


**Figure 3.** Internal friction loss as 1/Q versus H loading fraction in Pd at 114 K. Loss from H diffusion and dislocation motion are presented [4].

To summarize the significance these 3 experiments, the work of Syed (Fig. 2) simply proves that stable (or metastable) ~30% occupation of tetrahedral sites can exist at room temperature in PdDx and that he surmises, without measurement, that the presence of D in these sites increases strain due to the 3x smaller volume of tetrahedral sites and this strain changes the electronic band structure to yield increased Tc. The internal friction work of Mazzola (Fig. 3) indicates that the dynamics of H diffusion, where H must pass through tetrahedral sites to jump to the next octahedral site, is greatest at a H/Pd loading of 0.6, and this agrees with the conjecture of Syed that this is why he gets the highest tetrahedral occupancy for intermediate loading when soaked in H at 300 C. Now, it is conjectured here that the PAC results of 43% higher strain for PdDx (Fig. 1) is the same strain surmised by Syed and internal friction also agrees with the conjecture of Syed as to why he gets maximum loading ai D/Pd of intermediate loading. So, these three materials science measurements are related and tell us potentially important microscopic information about the Pd hydride system. The fact that the PAC method involves nuclear decay, and the 30% occupancy of tetrahedral sites was found through superconductivity measurements have nothing to do with LENR, but the microscopic facts uncovered by the measurements are related to LENR.

How would these facts apply to the AHE? The Hypothesis is modified to: *The AHE requires a specific frequency optical* ***phonon*** *resonance that is only attainable when there is* ***significant D occupation of tetrahedral sites***.

## 3. Palladium

As stated in the introduction, it is hypothesized that phonons in Pd and Ni play a role in generating and sustaining the AHE. *The AHE requires a specific frequency optical phonon resonance that couples to electromagnetic radiation of the same frequency.* This phonon resonant frequency in $PdD_x$ is a complicated function of the D concentration (cannot get right frequency with H), defect concentration of the right type, interstitial site occupancy (*fraction of octahedral vs tetrahedral*), temperature, polycrystalline texture, surface micro-topology. Why is the AHE so difficult to produce? Because only a specific set of conditions outlined in the previous sentence will produce the effect and the *fraction of*

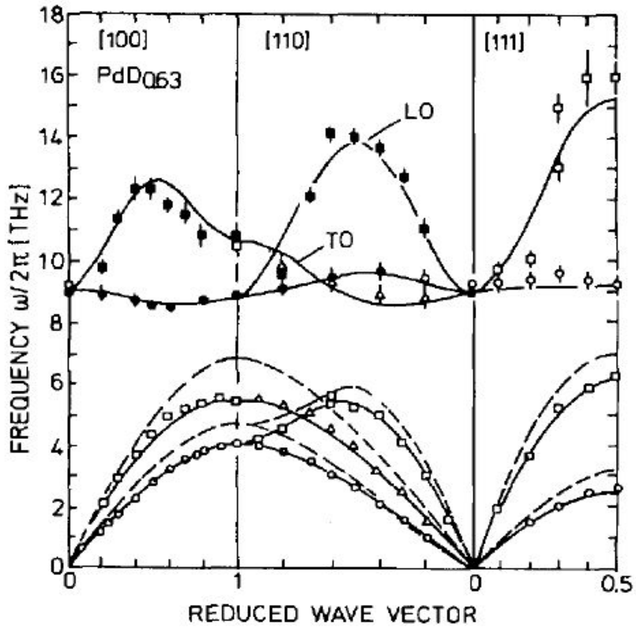


**Figure 4.** Measured phonon frequency dispersion versus major crystallographic axes in $PdH_{0.63}$ [6].

*octahedral vs tetrahedral* may be a critical one. Figure 4 shows the experimental optical and acoustic phonon spectra for $PdD_{0.63}$ [6] where phonon frequency in THz is the ordinate and the abscissa is different major axes in the crystal. Figure 5 takes the gamma <100> crystal direction and places a data point for the phonons on the plot for 6 different models of phonon frequencies [6], i.e., for Pd, $PdD_{0.63}$, PdH(oct), PdH(tet), $Pd_3VacH_4$(oct), $Pd_3VacH_4$(tet) where oct and tet stand for octahedral and tetrahedral sites, respectively, and Vac stands for vacancy. The first 2 are experimental and the last 4 are theory. Experiments can only be done for D and theory can only be done for H. Nevertheless, Figure 5 shows that the unique frequency could be over a wide range from 2 to 40 THz. In general, the addition of hydrogen increases the frequency of hydrogen sub-lattice modes and decreases the frequency of Pd sublattice modes.

Here it is conjectured that the special phonon frequency is not possible in Pd loaded with D where it is known that 100% is in the octahedral site, but promotion of significant D into the tetrahedral site will alter the phonon frequencies and one will overlap with the special frequency and heat is produced. Figure 5 demonstrates there is a second possible mechanism to achieve the special frequency – by insertion of vacancy defects into the Pd lattice. Keep in mind that the frequencies for any of the configurations will vary with changes to H concentration, temperature, crystal major axes. In this model, H will not produce the AHE because it's possible phonon frequencies never overlap with the required **specific frequency**. Figure 5 is not complete enough to try and pinpoint the specific frequency using the fact of exclusion of H and acceptance of D frequencies. Note that inelastic neutron diffraction found an 80 millielectron volt (20 THz) phonon in 8 nm $PdH_{.43}$ nanoparticles that is not in bulk PdH that is a candidate for the **specific frequency** [7].

The notion that a **specific phonon frequency** induces the AHE, leads to a straightforward mechanism that sustains the effect. Once triggered, the AHE mechanism itself produces either the right frequency phonon or the right frequency electromagnetic radiation that provides *the positive feedback to sustain the AHE mechanism*. A phonon resonance will

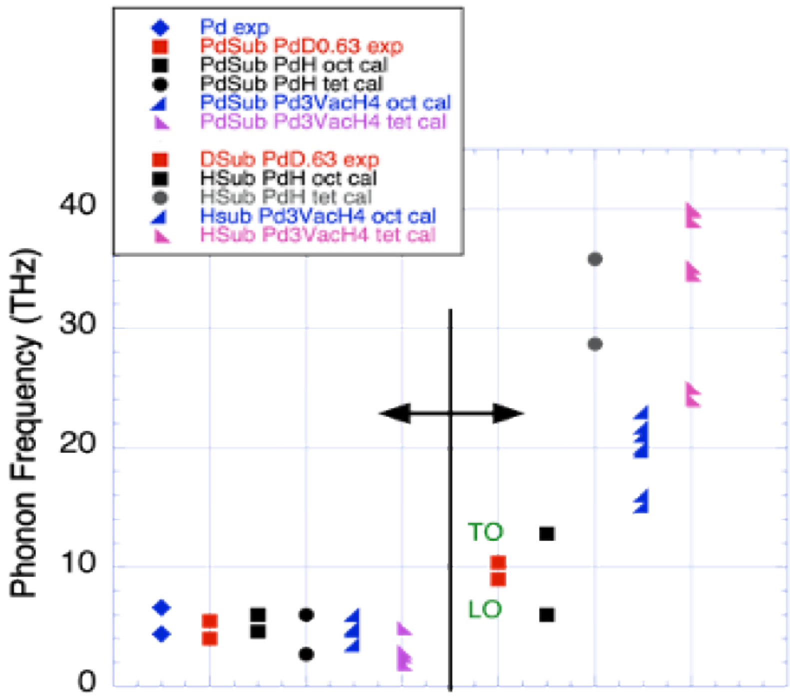


**Figure 5.** Phonon frequencies for the Pd and H sub lattices along the 100 axes for various configurations of H or D in Pd. Exp means experimental data and cal means calculated [6].

emit photons of the same frequency since the phonons are oscillating charges [8], [9], and electromagnetic radiation of the same frequency as the phonons will be resonantly absorbed, reinforcing the phonons.

Pd nanoparticles can play a role in producing the right frequency. It is calculated that 8 nm Pd nanoparticles have 30% occupation of the tetrahedral site [10]. The fact that nanoparticles may play a role dove tails well with experimental data. Start with Fleischman-Pons who often found that it took weeks before the excess heat effect began [11]. During electrolysis, the surface undergoes large modifications, dissolving Pd and reforming constantly and with incorporation of electrolyte impurities in the surface. Frequently there are deposits on the cathode surface after successful AHE runs. It is possible that over time, nanoparticles developed on the surface of the cathodes, and vacancies were inserted from the surface [12] and diffused in and caused the delay in the onset of the AHE. Figure 6 shows the deposits on a cathode that produced 730,000 Joules of excess energy over a period of 54 days [13]. Resonating nanoparticles on the surface from surface plasmon resonance or mechanical shape resonance, excited by charge exchange, will emit RF radiation and may not only cause the AHE in the nanoparticles but also stimulate phonons deeper into the bulk and engage a greater volume in the AHE process. Violante worked out in detail how nano-surface features on Pd could emit RF from MHz to THz [14].

Also, it was found that the addition of $\leq$ 9 nm Pd nanoparticles to an electrochemical cell increased the probability of observing excess heat [13]. Arata found the AHE using Pd nanoparticles in a heated, closed cell under $D_2$ pressure [15] as did Yan Kucherov in 1998 [16]. Cravens and Gempel used Pd and Au nanoparticles deposited on activated charcoal in $D_2$ atmosphere enclosed in a brass ball with a magnetic field [17]. The latter two

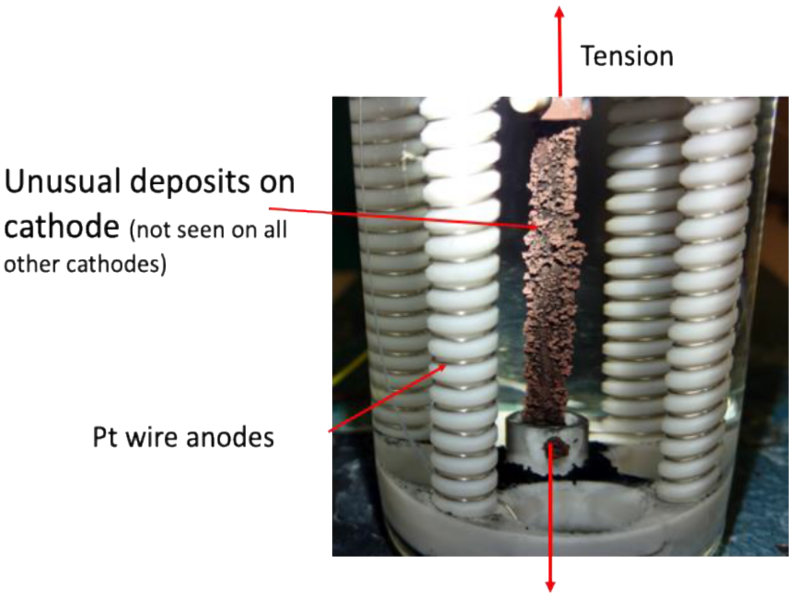


**Figure 6.** A Pd cathode that produced significant excess heat after 58 days of electrolysis showing deposits on surface [12].

emphasized a temperature gradient to encourage diffusion dynamics. Triggering can substitute for nanoparticles as shown by Staker [18] and Godes [19] who used electrical current pulses to stimulate the AHE in Pd rods. In this case electromigration could populate the tetrahedral site. D. Letts et al used lasers to generate THz radiation on the surface and obtained the AHE. The radiation could have provided the energy to populate the tetrahedral site with D [20]. Acoustic shock may promote D to the tetrahedral site. Energetics obtained the AHE using ultrasound stimulation [21]. It is sensible that both acoustic stimulation loading and glow discharge loading should be done gently since both may destroy triggering nanostructures formed on the surface. The polycrystalline morphology of the Pd cathodes can also play a role since a textured foil would benefit from phonon dispersion being the same across the volume of the foil or wire. This dove tails with the work of Violante who obtained evidence that the AHE is optimized for Pd highly textured in the 100 direction and for ~0.1 $\mu$m featured surface morphology [22], [23], [24]. To summarize this section, the special frequency is obtained by simple rules.

In bulk Pd

-a) Significant population of D in tetrahedral site (~30%), or
-b) High vacancy concentration
Either a or b provides the special phonon frequency that produces the AHE
-Deposit <10 nm Pd nanoparticles on surface

In nanoparticle Pd

- Use < 10 nm size
- Maximize volume of nanoparticles with access to deuterium
- Heat the nanoparticles

In general

- Stimulate this phonon to resonate by a trigger
- Sustain the stimulus to keep resonance active

*A 100 textured material will maximize heat (V. Violante) [25].

## 4. Nickel

Experiments using Pd are run below the Pd Debye Temperature and so phonons are possible excitations in the solid. For Ni, experiments are run at elevated temperatures that exceed the Debye temperature (450K) where phonons are not possible. However, the boundary conditions extant in nanostructured materials recover the ability of Ni-based materials to support phonons and phonon-like, size-based excitations at elevated temperatures [7]. Ni-based AHE embodiments all use nanostructured materials and Ni and Pd share the same crystal structure and similar band structure.

The experiments of Parkhamov (Ni powder, $LiAlO_2$) [26], Celani (NP coated constantan wires) [27], Swartz ($ZrO_2PdNi$ powder) [28], Godes (Ni wire) [19], Kitamura (Ni powder) [29], Mizuno (Ni mesh) [30], Focardi (Ni powder) [31] all use Ni-based nanoparticle formulations in their reactors or coated wires. Iwamura [32] and Kasagi [33] use nm thin-film multilayers containing Ni. In the case of Ni:

- Nanostructures are preferred over bulk materials on the order of 10 nm. This may be due to the facts that experimental temperatures exceed the Debye temperature and the very low solubility of H in Ni
- Since temperature thermally drives the phonons, there is no need for a trigger
- Maximize volume of nanoparticles with access to hydrogen
- Since H produces heat, the $NiH_x$ nanostructures produce the **special** phonon frequency
- AHE with Ni is more reproducible than with Pd (no need to kick D into T-site or produce high a vacancy concentration and the trigger is simply thermal).

## 5. Discussion

This model was developed by noticing that nanostructured materials are a commonality among different AHE methods, by noting experiments that exposed real microscopic differences between PdH and PdD, and by the application of common-sense organization to the data. For Pd, nanoparticles play a role of generating excess heat themselves and in addition serve to excite bulk phonons if the bulk phonons are prepared with some fraction of tetrahedral occupation or vacancy concentration. Only loading with D results in the AHE. For Ni-based materials, nanostructured materials seem to be active and bulk materials less so. Loading with H will result in the AHE and perhaps with D, but this is not well established. While this phonon resonance model seems to unify the many different AHE results, *this resonance-phonon model is agnostic as to what the actual AHE mechanism is.* Therefore, the physics of the actual AHE energy source must be mapped onto this model by devising either a source of specific frequency E&M radiation or a mechanism to drive energy into atomic motion of specific frequency phonons, keeping in mind that the only signatures of the AHE are *heat production*, *RF emission*, *no nuclear emissions.*

Some lessons from this model to guide experiments are:

- Work with Pd nanoparticles with size 9 nm or less
- For good hydrogen dynamics, stay below D/Pd $\leq 0.64$
- Triggering of Pd to promote D to the tetrahedral site helpful – electric pulse has been demonstrated
- Use highly textured Pd – 100-orientation preferred – likely for Ni also Work with Ni nanostructures with size 14 nm or less. Much less determined than for Pd

- Ni does not need trigger – just heating is enough
- Glow discharge and ultrasonic loading should be done gently so as not to destroy surface nanofeatures
- Model implies that the AHE is a bulk phenomenon, not a surface phenomenon.
- The Ni-based materials at elevated temperature yield more reproducible AHE results than $PdD_x$ materials in electrolytes.

Some suggestions for future fundamental experiments include:

- Perform the internal friction measurement for D loading in Pd instead of H and repeat the loading/deloading cycle several times on the same specimen.
- Examine the Tc = 61 K $PdD_x$ material in an AHE apparatus with triggering
- Characterize RF emissions
- Diffuse radioactive $^{57}$Co into Pd foils or Ni nanostructures and perform Mossbauer spectroscopy during excess heat events. Look for changes in decay rate or energy shift when producing heat (injects nuclear physics into experiment at 14 keV level)
- I recommend the Iwamura-Kasagi method since it is done in hard vacuum where many diagnostics become possible [32], [33]
- Add 2% Ni to Pd which renders Pd Ferromagnetic and do electrolysis with magnetic field
- Place Pd and/or Ni in fast neutron flux from a nuclear reactor to introduce high concentration of vacancies and perform AHE experiments.

A word about the well-known equation reported by McKubre which states that to obtain the AHE the D loading in Pd must be >0.88 and that the excess energy is proportional to $[X – 0.88]^2$ where x is the deuterium concentration D/Pd [34]. Also, that the current density must be 250 to 500 mA/cm$^2$. This is huge current density, causing a region of dynamical instability near the surface. While electromigration may populate T sites, vigorous electromigration will also depopulate the T sites and higher temperature of the cathode due to IR heating also reduces stability of D in T site. We speculate that for a cathode that will readily load to D/Pd >0.9, the large fugacity at 500 mA/cm$^2$ still forces D into the surface so when D/Pd gets to ~0.88 then D jumps to T site and has fewer paths to depopulate so concentration in T site begins to grow (see Fig. 1). Once this happens, the T-site population will grow as the square of the concentration above 0.88 which is what McKubre observed.

The anomalous heat effect is, to first order, a fundamental a solid-state physics problem – not a nuclear physics problem. There are unknown excitations occurring in the solid that produce heat. A survey of the several hundred named solid-state phenomena provide no clues. We need measurements that reveal phenomena at the scale of the atom in relevant material systems as represented in this work. This is a bottom-up approach where we search for the unknown excitations and then try to map them onto an energy generating process.

Finally, I offer suggestions that may lead to a mechanism. Hagelstein's theory is phonon based where a particular optical phonon in the few to 40 THz region of frequency undergoes coherent resonance and couples to virtual 2 nucleon energy levels and the energy from fusion to $^4$He is dissipated by ~E8 atoms in the phonon field, and the mechanism solves the problem of the lack of energetic emissions [37]. The model presented here dovetails with Hagelstein's theory. Another place to look for an excitation that may relate to the AHE mechanism is as follows. Density Functional Theory (DFT) teaches us that the electronic band structure is *very sensitive to the interatomic spacing*. Imagine two atoms in an optical phonon at the extremity of excursion away from their equilibrium positions (equivalent to strain). For the few femtoseconds at the extreme position, an entirely different electronic band structure can evolve due to a different atomic spacing. A few fs is a long time from the electron's point of view. We can have metal to insulator, metal to semiconductor, new superconductor, non-ferromagnetic to ferromagnetic, etc., transitions [35], [36]. Such a dynamic effect is difficult to explore experimentally and may require fs timing methods. To emphasize this point, the

Tc of 61 K in PdDx and the 50% increase in Tc in $MgB_2$ mentioned above are both caused by static tensile strain that changes the interatomic spacings. These nonequilibrium electron configurations may cause, for example, colossal crystal fields to momentarily emerge.

The struggle to uncover the mechanism(s) that produce the anomalous heat effect continues. Perhaps the insights described in this paper will help in that endeavour.

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