Second-Order Quantum Coupling Effects and Energy-related Problems

Academic and Research Staff
Professor Peter L. Hagelstein

Collaborators
Dr. Yan Kucherov[1], Dr. Graham Hubler[2], Dr. Mike McKubre[3], Dr. F. Tanzella[3], Dr. Andrew Meulenberg[4] Robert DiMatteo[4], Prof. Clifton Fonstad

Visiting Scientists and Research Affiliates
Dr. K. P. Sinha

Graduate Students
Irfan Chaudhary, M.M. Masaki

Support Staff
Susan Chafe

Introduction

Energy issues have become increasingly important for our world. It could be plausibly argued that energy will shortly be the single most important problem for industrialized countries, on the same footing as food, water and overpopulation, for the world as a whole. On a timescale of decades, our world may face an inevitable exhaustion of nonrenewable energy supplies. Even at the present time, energy issues weigh heavily in political events around the globe. Instability in the Middle East has the potential to lead to large increases in energy costs in the US, which may help to focus the attention of this country on a problem that has been important for some time.

The research described in this article is concerned with three different energy-related research topics. Two of these are conventional and one is controversial. The first topic concerns an advance made at Eneco, Inc., which recently received some attention in the national and international media concerning the development of new devices, which enhance the ability of a thermoelectric semiconductor to convert heat to electricity. The associated technology which is anticipated to result from this advance will lead to commercial and military solid state devices that convert low-grade (100-500C) heat to electricity more efficiently than conventional thermoelectric devices.

The second topic concerns a basic physics experiment that involves the possible application of a second-order off-resonant quantum coupling effect to microcavity thermophotovoltaics. Thermophotovoltaics is anticipated to become an important technology for the conversion of high-grade (500-2000C) heat to electricity for military and commercial applications. Microcavity thermophotovoltaics improves on conventional thermophotovoltaics by increasing the power density per unit area throughput. The new proposed quantum-coupling experiment that we discuss has the potential to provide a further increase in the energy throughput, and in the efficiency associated with the excitation transfer effect (being operationally equivalent to having a narrow-band source).

The third topic has been controversial for the past 13 years, and involves the observation of anomalies in metal deuterides, and research devoted toward trying to understand what physics underlies the basic

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[1] Eneco-USA, Salt Lake City, UT
[2] Naval Research Laboratory
[3] SRI International, Inc.
[4] Draper Laboratory

effects. The situation as it stand today is more or less as follows: Many experiments on metal deuterides exhibit a variety of anomalies. The experiments are consistent with the existence of new and unexpected nuclear effects. Due to the controversy of previous years and negative publicity, most scientists doubt that there exist any real anomalies, associated research and public discussion is not encouraged, and publications are generally discouraged. From our perspective, the experimental results seem real enough, and the important outstanding question concerns what physical mechanisms are responsible. This research area might be considered under the general heading of "emerging" science.

One of the anomalies is the claimed observation of an excess heat effect, and the associated quantitative production of ^{4}He. If it is true that deuterium can be "burned" to produce a ^{4}He ash, then a door opens to a new kind of nuclear energy technology. This is of course a major goal of interest to scientists working in this field. Much controversy in previous years centered on the relative lack of helpful experimental results and the complete absence of any reason to hope for the existence of a physical mechanism which might allow such a process to exist. In the end, the underlying questions are physics questions, which will ultimately be answered in the laboratory. Over the past several years, a variety of experimental results and theoretical ideas have led to the consideration of a new model, which appears to have the potential to make sense out of the seemingly disparate anomalies. Our discussion in the third section is intended to provide an overview of the situation in the field in light of the new model as it is presently understood.

1   Enhancement of the Efficiency of a Semiconductor Thermoelectric

Project Staff
Peter Hagelstein and Yan Kucherov

When a temperature gradient is imposed across a conductor, in most cases a small voltage will be induced. If the conductor is shorted out, a current will flow. One might think of the origin of the current in terms of carriers within the conductor moving on average away from the hotter region of the conductor in the general direction of the colder region. To develop zero total current as required in the case of open circuit conditions, we might think of the thermally-induced current just described as being balanced by an ohmic return current. The build-up of net charge within the conductor gives rise to an internal field, which drives a return current in the opposite direction. This is observable as an open circuit voltage. The development of a voltage and current in the presence of a temperature gradient in this way constitutes the thermoelectric effect.

Thermoelectric materials are used for thermal to electric energy conversion applications and for refrigeration. The goodness of a thermoelectric material depends on its ability to develop a voltage (which is characterized in terms of the thermopower Σ, which is a measure of how much voltage per unit temperature drop), the electrical conductivity σ and the thermal conductivity κ. How good a thermoelectric material is can be determined from its thermoelectric figure of merit. In the literature, thermoelectric materials are often characterized in terms of the *ZT* parameter

$$ZT = \frac{\Sigma^2 \sigma}{\kappa} T$$

The three material parameters Σ, σ, and κ are functions of the thermoelectric material under consideration. In most cases, there is little that one can do to a material that will have much effect on these parameters so as to improve the figure of merit. During the last few years, materials scientists have developed a variety of interesting new approaches to designing materials with special properties, which result in modifications of the material parameters. For example, phonon transport can be inhibited through the use of layered materials. The thermopower and electrical conductivity can be improved in

thin films and in nanowires. For n-type thermoelectric materials, the largest *ZT* parameter reported so far in the literature is in the neighborhood of 2.5.
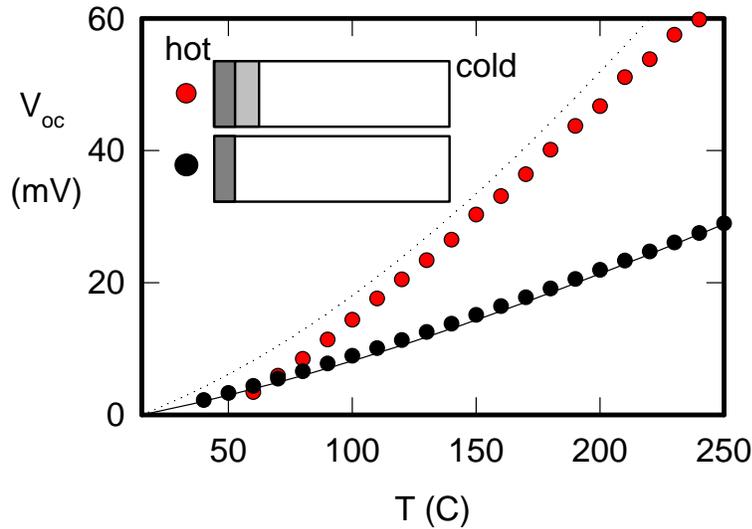
The thermal diode

We have recently found a way to improve the efficiency of semiconductor thermoelectric materials for thermal to electrical energy conversion, through the use of a new device, which we have termed a thermal diode [1,2]. In the simplest implementation, the thermal diode consists of a thin (micron-scale) emitter layer on the hot side of a thick (millimeter-scale) near-intrinsic semiconductor thermoelectric (see Figure 1.1). We have found that the emitter layer injects a current into the bulk thermoelectric region. The short-circuit current density is found to increase by a factor of two or more, which we attribute to current injection from the emitter. The open-circuit voltage is also observed to increase by a factor of two or more, which we attribute to the greater internal field that is required to produce a larger ohmic return current.

Figure 1.1 Basic thermal diode structure. Second-order thermionic current injection from the heavily doped emitter diffuses across the solid gap region to the collector. The solid gap region is a near-intrinsic thermoelectric semiconductor, which contributes a thermoelectric current in addition to the injected current.

The mechanism, which we have proposed to account for this effect, is a new kind of second-order thermionic emission process.   The scattering lengths for electrons in the materials that we have considered so far (indium antimonide and mercury cadmium telluride) are very long (on the order of a micron). We consider briefly the current injection under short circuit conditions. An electron can transport ballistically from the emitter across the junction into the edge of the hot side of the bulk thermoelectric material. One might view this injection as a thermionic effect, but there is a similar reverse injection in the other direction that very near balances the forward injection. If the electron distributions were assumed to be in equilibrium on both sides of the junction, then the forward and reverse currents would very nearly balance (there would be a net thermoelectric current consistent with the conventional thermoelectric current associated with a layered device). However, as we are dealing with thermoelectric materials, the electron distributions have a non-Fermi-Dirac contribution, and the ballistic transport of the non-Fermi-Dirac part of the distribution does not lead to a balance. The net result according to such a model is that there can be an injected current in addition to the thermoelectric current, and which can be up to about four times as large in theory as the thermoelectric current. The largest enhancement seen so far in experiments at Eneco is a factor of 2.8 increase.

Figure 1.2 Open-circuit voltage as a function of current for an InSb thermal diode with a poor emitter (black solid circles); thermoelectric model open-circuit voltage (solid line); open-circuit voltage of a thermal diode with a good emitter (red solid circles); model open-circuit voltage calculated from the current injection model (dotted line). Current injection from the emitter has increased the open-circuit voltage by a factor of 2.2 over the thermoelectric result.

**Enhancement of the open-circuit voltage and effective figure of merit**

An increase in the open-circuit voltage implies an enhancement in the associated figure of merit parameter. A measure of the improvement in performance of the device over the thermoelectric performance can be developed directly from the experimental measurements through the definition of an effective figure of merit parameter. We have defined the effective integral figure of merit $Z_{eff}T$ parameter in terms of the experimental observables to be

$$Z_{eff}T = \frac{J_{sc}V_{oc}}{Q_0 \Delta T}T$$

Here, the short-circuit current is J_{sc}, the open-circuit voltage is V_{oc}, the zero-current heat flow is Q_0, and the temperature drop across the sample is ΔT. In the Eneco experiments, the heat flow is approximately linear in the temperature drop. The current and voltage relation is approximately linear (with an offset). Consequently, the effective integral figure of merit for the thermal diode is improved over the equivalent thermoelectric figure of merit by the ratio of the voltage increase squared.

$$Z_{eff}T = \left[\frac{V_{oc}^{TI}}{V_{oc}^{TE}}\right]^2 ZT$$

A voltage enhancement of a factor of 2.8 corresponds to an increase of a factor of 8 in the effective integral figure of merit over the thermoelectric value.
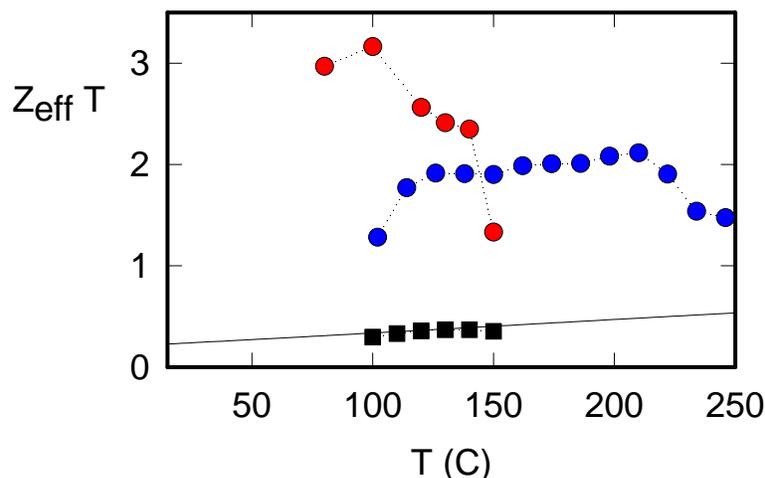
Figure 1.3 Experimental results for $Z_{eff}T$ for a mercury cadmium thermal diode (high solid red dots); a layered device with an InSb thermal diode on the hot side, and a HgCdTe thermal diode on the cold side (intermediate solid blue dots); a copper-cladded thermoelectric HgCdTe device (solid black squares); thermoelectric model result (solid line). Results are given as a function of the hot side temperature. The effective figure of merit $Z_{eff}T$ has been increased by about a factor of 8.

We have also demonstrated that the ohmic return current can be inhibited, by placing a compensation layer on the cold side of a weakly doped thermoelectric semiconductor next to the collector contact. An enhancement of the effective figure of merit on the order of a factor of two has been observed in different experiments as a consequence of this effect.

Thermal diodes have been implemented in indium antimonide, and in several mercury cadmium telluride alloys (with different cadmium fractions), with enhancements in the figure of merit being in the range of 5 (InSb) to 8 (HgCdTe). The highest $Z_{eff}T$ product that we have seen is on the order of 3.2, starting from a near-intrinsic mercury cadmium telluride sample for which the thermoelectric figure of merit ZT was 0.40. We believe that similar enhancements can be seen in other semiconductor thermoelectric materials. The results to date suggest that it may be possible to develop a total enhancement on the order of 20 with an optimal thermal diode design and implementation.

[1] P. L. Hagelstein and Y. Kucherov, "Enhancement of thermal to electric energy conversion with thermal diodes," *Proceedings of the MRS Winter 2002 Conference*, Boston, Nov. 2001.

[2] P. L. Hagelstein and Y. Kucherov, "Enhanced Figure of Merit in Thermal to Electrical Energy Conversion with Thermal Diodes," to appear in *Appl. Phys. Lett.*

[3] Y. Kucherov and P. L. Hagelstein, "Efficient thermal to electrical conversion using thermal diodes," *Bull. APS* (March Meeting 2002), **179** (2002).

[4] P. L. Hagelstein and Y. Kucherov, "Enhancement of the figure of merit for thermal to electrical energy conversion with thermal diodes," *Bull. APS* (March Meeting 2002), **179** (2002).

2   Power Enhancement from Quantum Coupling between Semiconductors for Photonic Based Energy Conversion

Sponsors
Draper Laboratory

Project Staff
P.L. Hagelstein, K.P. Sinha, A. Meulenberg, R. S. DiMatteo, C.G. Fonstad

ThermoPhotoVoltaics (TPV) are a class of photonic based electrical generation devices. TPV devices convert heat to electricity by heating a surface (the emitter) which radiates IR photons to an adjacent low band-gap photovoltaic cell (typically InGaAs, GaSb, InAs, or InGaAsSb) which converts these IR photons to electricity. It was hypothesized [1] and recently demonstrated [2] that, if the hot emitter surface is brought into submicron distance from the cooled photovoltaic cell, substantially increased photonic and electromagnetic flux from hot side to cold side will occur - thereby increasing the photovoltaic output. The basic effect in this case is purely classical, due to additional power flow in the evanescent modes. A rapidly growing body of literature has been providing further theoretical understanding of these devices with this micron and submicron vacuum gap between hot and cold side and has been referred to as Micron-gap or Microscale TPV (MTPV).

Possibility of quantum coupling

We have been interested over the last several years in the possibility of finding physical applications for a novel second-order quantum coupling mechanism that has been described in previous RLE Progress reports. The basic idea is that excitation can be exchanged between two-level quantum systems when both are coupled to a common oscillator. If the oscillator is off-resonant, then the basic effect is an example of a second-order off-resonant quantum-coupling mechanism. We have described previously how odd such an effect might appear. In what we described previously [3]., we might imagine atoms in one side of a microwave cavity that are excited, but allowed only to couple to a common microwave field. On the other side of the microwave cavity are identical atoms, which differ only in being unexcited, but also coupled to the microwave cavity. If the interaction between the atoms and the microwave field is somehow made to be nonlinear, then it is possible in principle to transfer the optical energy from one set of atoms to the other through coupling with a microwave oscillator.

Here we consider an application of the same second-order off-resonant coupling mechanism to quantum-well states in closely spaced semiconductors. We imagine that the quantum-well levels on either side are well approximated by two-level quantum systems. We recognize that there exists resonant coupling through transverse-photon exchange, which can be a near-field effect if the two quantum wells are within a wavelength or closer. We recognize that there can also exist resonant evanescent-wave effects, again through coupling with transverse photons.

In addition, we anticipate the possibility of coupling through a variety of off-resonant channels. We expect off-resonant coupling through transverse-photon exchange at frequencies different from the resonant energy of the two-level systems. Longitudinal photons have zero energy in QED, and any coupling via longitudinal photon modes will constitute an example of off-resonant second-order coupling. These processes could dominant in the case of very closely spaced quantum wells separated by a vacuum gap. In our proposals for an initial demonstration of the effect, it has proven convenient to work with a solid gap between the quantum wells. This is because it is relatively difficult to maintain macroscopic semiconductor quantum-well structures at a vacuum spacing on the order of a few hundred Angstroms. Consequently, there exists the possibility of phononic coupling between the states in the two wells. Any transfer of optical excitation through phononic coupling is decidedly an off-resonant coupling effect.

In the event that the off-resonant effects dominate over resonant-coupling effects, then we will have demonstrated, perhaps for the first time, an implementation of an off-resonant second-order quantum-excitation transfer effect. If the effect is found to work in the presence of a small vacuum gap, then it may have ramifications for the energy conversion problem. For example, in the event that the off-resonant effects become dominant in this case, we will see an enhancement of the excitation transfer from one side to another over what is possible through classical electromagnetic coupling. As we have both theory and experimental data for the classical coupling problem, we will be able to determine whether the new quantum coupling effect is operative. It is predicted to show up as an enhancement of the coupling, which implies an improvement in the efficiency of small gap TPV device operation through this mechanism.

Design issues

To implement the scheme, we require a quantum-well design and an associated analysis. In the case of classical evanescent wave effects, extensive modeling has been done. Mathematical models developed at MIT [4] and Draper Lab [5], dispense with the details of the mechanisms. They use "average" material properties to identify resonances and then to predict the proximity-enhancement effects. Figure 2.1 provides an example of the Draper Lab electromagnetic-theory enhancement-model results [6]. It shows the output power density for 2 μm light from a 1000K silicon emitter into an InGaAs TPV detector. The portion of the curve above 10^{-7}m is the physical-optics-coupling (n^2 effect) enhancement. The enhancement here is not limited to n^2, since other optical-loss mechanisms are also removed by reducing the gap toward zero. [Antireflective coatings on the emitter and detector would raise the far-field power coupling; but, they would also reduce the microgap coupling.] The portion of the curve in Figure 2.1, below 10^{-7} m, indicates further enhancement from the emitter/detector interaction. This model probably does not adequately address the new quantum-coupling effects. Nevertheless, it does indicate a significant proximity enhancement beyond the n^2 effect.
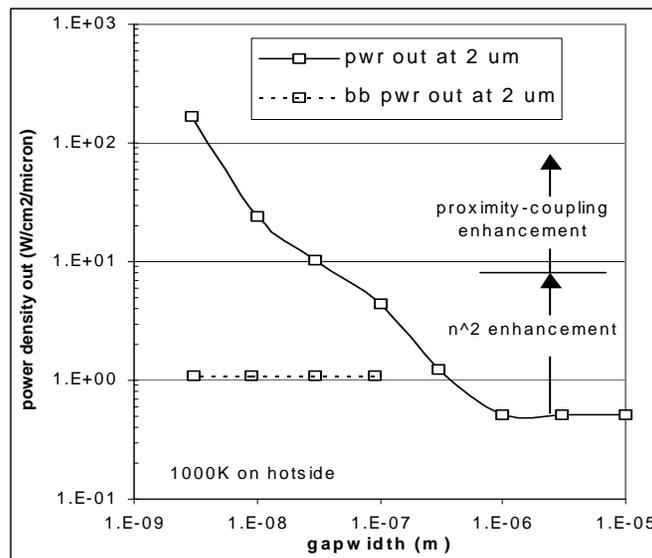

Figure 2.1. Electromagnetic theory enhancement model results. Output power density for 2 um light from a silicon emitter into an InGaAs TPV detector.

Some work has been done attempting to develop design and analysis capability for the quantum-coupling part of the problem in particular. In a realistic design, one might like to include various complicating

effects, such as the presence of a continuum of transverse photonic modes, longitudinal photonic modes, phononic modes, line broadening effects, and many-body effects.

Some initial results from the MIT quantum mechanical model have recently become available. Figure 2.2 includes the quantum-coupling enhancement, along with the n^2 effect, for a proximity-coupled 660K InGaAs emitter into an InGaAs TPV detector. Note that, in this case, the enhancement exceeds 10x between 5×10^{-7} and 10^{-7} m. But, even more important, the power transfer is comparable to that for the silicon emitter (represented in Figure 2.1) at 1000K. This large enhancement is predicted for discrete-state to discrete-state coupling. However, the line-broadening effects are significant (corresponding to a single-sided half-width at half-maximum, HWHM = 0.05, of a direct-bandgap semiconductor). The line broadening of a higher-temperature emitter will be somewhat greater and, therefore, will further broaden and lower the peak enhancement in Figure 2.2. The effect of the continuum of states above a semiconductor band edge has not yet been quantified. However, it is expected to raise and broaden the peak above 0.6 eV. Understanding and quantifying this effect is one of the goals of the experiments presently being undertaken.
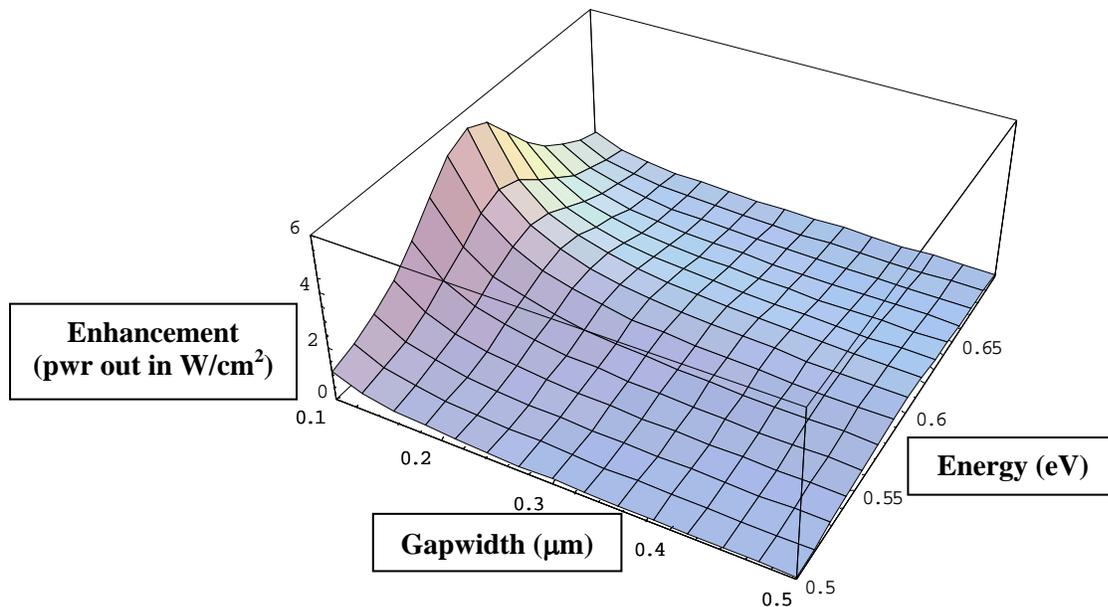

Figure 2.2 An example of proximity enhancement for resonant states at 0.6 eV. Effects of gapwidth and line broadening are displayed.

Initial experimental proposal

The proximity coupling of photons across a vacuum microgap has been experimentally confirmed [2]. The new quantum coupling effects at close spacing will be identified in an experiment that uses a dielectric gap and a light source, to generate emitting electrons, rather than a thermal source. Figure 2.3 is a diagram of a semiconductor quantum-well structure proposed to verify and quantify the proximity-coupling effects beyond the n^2 effect. Since, there is no vacuum gap between the emitter and detector, the n^2 effect will be negligible. The multiple layers of the test structure will be grown by molecular-beam epitaxy, MBE, and thus can be made very thin (in the nanometer range) and well controlled. In this manner, the dielectric "gap" (or window) can be made much thinner than possible with a vacuum gap. Therefore, it is possible to vary and probe the ultrathin gap thicknesses plotted in Figure 2.1.

The structure will be grown on an InAlAs substrate. The first layer grown will be InGaAlAs with a bandgap low enough to absorb 1.3 μm light that would pass through the substrate. This absorbed light

would generate carriers that would be collected in the next layer, an InGaAs quantum well. The thickness of this well will be adjusted to provide a ground-state-transition level at ~0.855eV (~1.45μm). The next layer is the InAlAs window. Several versions of this structure will be made that are identical in all respects except for the thickness of this window. The fourth layer will be another InGaAs QW layer. In this case, the layer will be thicker, so that the second energy-transition level will be at the same energy as the light radiated from QW #1. Light absorbed in QW #2 (from QW #1) will generate electrons in a level from which they can decay/relax to the lower level before recombining to generate light at a third energy. This light emitted from QW #2 will have a longer wavelength (~1.5 μm) than that of the 1.3 μm light from the source and the ~ 1.45 μm light from QW #1). The final InAlAs "top cap" layer will establish the boundary of QW #2.
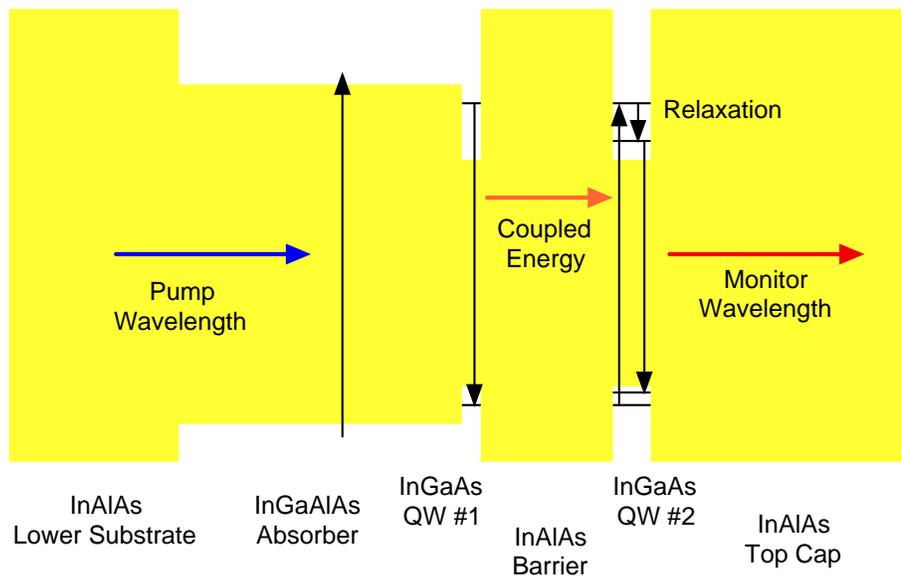
Figure 2.3. QW test structure to probe proximity and resonance coupling.

A spectrometer, looking at the sample, can then distinguish the 3 primary light sources. The light intensity from QW #2 and the ratio of the light emitted from QW #1 to that from QW #2 will change with window thickness. This information will be used to determine both the proximity- and the resonance-enhancement effects. Subsequent tests with an appropriate InGaAlAs layer between QW #2 and the top cap will allow exploration of a band edge (rather than a bound-state) coupling with QW #1 and absorbing its radiation.

Microgap experiments

Another experiment, similar to that in Reference [2], will be performed with matched semiconductors across a microgap. Using a lattice-matched InGaAs layer on InP substrate, as an emitter, and a lower-bandgap InGaAs layer (on InP), as the detector, it is possible to see the effect of resonance at a bandedge. As the emitter is heated to higher temperatures, its bandgap will shrink and approach that of the detector. As the emitter band edge crosses over that of the detector, the ratio of microgap to wide-gap photocurrents should trace out the curves of Figure 2.2.

The enhancement in photocurrent of this experiment relative to that in Reference 2 will indicate the extent of the proximity effect for matched (selective) emitter/detectors. The peak height and shape will indicate the line-width effects of a band edge on the coupling.

[1] DiMatteo, R.S., *Enhanced Semiconductor Carrier Generation Via Microscale Radiative Transfer*, Masters Thesis, Department of Electrical Engineering and Computer Science, and Technology and Policy Program, Massachusetts Institute of Technology, June 1996.

[2] DiMatteo, R.S., P. Greiff, S.L. Finberg, K.A. Young-Waithe, H.K H. Choy, M.M. Masaki, C.G. Fonstad, "Enhanced photogeneration of carriers in a semiconductor via coupling across a non-isothermal nanoscale vacuum gap", *Applied Physics Letters*, **79**, No. 12, September 17, 2001, p. 1894.

[3] Hagelstein, P.L., "Off-Resonanct Superradiant Coupling between Atoms and a Cavity," *RLE Progress Report* **141**, 1998, Part 1, Section 3, Chapter 1

[4] Sinha, K. P., "Quantum Theory of Radiative Transfer Between Closely Spaced Surfaces," Contract Report, in Progress

[5] Weinberg, M., "MTPV Solutions Over Many Layers," Draper Lab Memo, GBB4-01-0359, Dec. 31, 2001

[6] Meulenberg, A., "Separation of Optical and Near-Field Effects in MTPV," Draper Lab Memo, GBB-02-draft.

3   Anomalies in Metal Deuterides

Personnel
Prof. P. L. Hagelstein, Dr. G. Hubler, Dr. M. McKubre, Dr. F. Tanzella, Dr. K. P. Sinha, I. Chaudhary

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During the past 13 years, observations of a variety of anomalies from metal deuterides and hydrides have been reported. Under discussion during the past view years is the conjecture that many of the anomalies can perhaps be accounted for from a single underlying new physical process. Various proposals for what physics might be involved has changed over the years as we learn more about the experiments and about the models.

In what follows we outline the basics of the associated "unified" model as we understand it today. The model is considered to be unified in the sense that it appears to apply to almost all of the anomalies systematically. No other model can make such a claim. Following the development of a brief account of the model, we then provide a connection between the model and many of the specific anomalies that have been reported.

The model itself is ultimately a consequence of including solid state effects at the outset in the description of nuclear fusion reactions in a lattice. The inclusion of the lattice allows for phonon exchange between the macroscopic lattice and the microscopic nuclear system. Phonon exchange can mediate the transfer of angular momentum from the lattice to the nuclei, and also provides the underlying coupling for new interactions between the nuclei and the lattice. Most important of these new interactions is the possibility of second-order site-other-site reactions between nuclei at different sites interacting with a common phonon mode. The results of this interaction lead directly to observable consequences, which we discuss below.

The premise: Including solid state effects in nuclear reaction models

The premise of the model under discussion is that when describing nuclear reactions that occur in a solid-state environment, the solid-state environment should be included in the theoretical description at the outset. This we recognize as a radical proposal, with important consequences. In this subsection, we are concerned with the details of the adaptation of a model for fusion from the nuclear physics literature to be generalized so as to include lattice effects explicitly at the outset.

The inclusion of the lattice can be incorporated in the case of fusion reactions through appropriate generalizations of the resonating group method [1] or the R-matrix method [2]. We consider the resonating group method briefly. We might think of the resonating group method as developing a total wavefunction Ψ_T for the reacting channels in vacuum of the form $\Psi_T = \Sigma_j \Phi_j F_j$, where the Φ_j account for the internal nuclear wavefunctions, and the F_j are the channel separation factors. Optimization of the channel separation factors leads to coupled channel equations for the channel separation factors, which we write formally through

$$ \mathrm{E}\, F_i \;=\; \langle\, \Phi_i \mid\, \mathrm{H} \,\mid \Phi_i \rangle\, F_i \;+\; \sum_{j \neq i} \;\; \langle\, \Phi_j \mid\, \mathrm{H}\text{-}\mathrm{E} \;\mid \Phi_j\, F_i \,\rangle $$

The generalization of this to the lattice resonating group method involves the replacement of the channel separation factors F_i with lattice channel separation factors Ψ_j which keep track of the center of mass coordinates of the reacting nuclei as well as that of neighboring nuclei. The total wavefunction Ψ_T is now generalized to include lattice effects $\Psi_T = \Sigma_j \Phi_j \Psi_j$, and the lattice channel functions are optimized according to

$$ \mathrm{E}\, \Psi_i \;\;=\; \langle\, \Phi_i \mid\, \mathrm{H} \,\mid \Phi_i \rangle\, \Psi_i \;+\; \sum_{j \neq i} \;\; \langle\, \Phi_j \mid\, \mathrm{H}\text{-}\mathrm{E} \;\mid \Phi_j\, \Psi_j \,\rangle $$

We see that the generalization of the resonating group method, which was important early on in the nuclear physics literature for the analysis of the dd-fusion reaction, is accomplished without difficulty. This generalization, or the equivalent generalization of the R-matrix method, constitutes the underlying physical theory on which further discussion and developments are based.

Phonon exchange

One consequence of the new model is the possibility of phonon exchange during a fusion reaction with the lattice. Within the lattice resonating group method, the strong force interaction potential can be viewed as a nonlinear phonon operator. For example, the center of mass position operator $\hat{\mathrm{R}}_k$ of a nucleus can be developed in terms of phonon mode operators $\hat{\mathrm{q}}_m$ through an expansion of the form

$$ \hat{\mathrm{R}}_k = \mathrm{R}_k^{(0)} + \sum_m \mathrm{u}_m[k]\hat{\mathrm{q}}_m $$

The nuclear interaction between nucleons of reacting nuclei in a pion exchange picture now must be thought of as being a highly nonlinear phonon operator [3]. To illustrate this, one might envision one of the nuclear potential models (for example, such as the old Hamada and Johnston potential [4]) as constituting an enormously complicated function of the phonon $\hat{\mathrm{q}}_m$ operators, when taken between nucleons associated with different nuclei

$$V(r_2\text{-}r_1) \ = V(\Delta\,\hat{R} + \mathbf{x}_2 - \mathbf{x}_1) \ = \ V(\Delta\mathbf{R}^{(0)} + \mathbf{x}_2 - \mathbf{x}_1 + \sum_m \Delta u_m[k]\hat{q}_m\,)$$

where the nucleon position $\mathbf{r}_i$ is assumed to be related to the nuclear center of mass position as $\hat{R}_i + \mathbf{x}_i$.

We see in this is that since the relative positions of the center of mass of different nuclei depend explicitly on the amplitudes of the lattice phonon modes, that the interaction between the nucleons of one nucleus and the nucleons of a different nucleus are functions of the phonon mode amplitude operators. Consequently, phonon exchange is expected to occur between the lattice and nuclei in the course of a fusion reaction between neighboring nuclei in the lattice.

As the Coulomb barrier normally keeps nuclei well apart from one another in the lattice, the issue of phonon exchange is normally not an issue. If a deuteron beam is incident on a metal deuteride at tens of keV, one would not expect the result of a theoretical calculation to be much different if a few phonons are exchanged or not. The reaction energy could change, but the effect would be at most tens of meV as compared to several MeV of reaction energy.

Phonon exchange during a fusion reaction has the potential to modify the microscopic selection rules. For example, the exchange of phonons during a dd-fusion reaction could allow the ^{4}He channel to be accessed (assuming that two angular momenta quanta are exchanged in association with the phonons) as an off-resonant virtual channel. It is impossible for two deuterons to react directly through phonon exchange to make ^{4}He since an exchange of at least 500 million optical phonons would be required, and the nonlinearity of the nuclear force is not strong enough to mediate such an exchange.

Second-order two-site reactions

Reactions that occur at different sites can become coupled if they exchange phonons with a common phonon mode. If coupling occurs between two exothermic reactions, whatever effects might show up at second order would likely be impossible to observed as they would be small. A coupling between and exothermic reaction at one site and an endothermic reaction at a different site can be a different story.

The most important two-site reaction process in a metal deuteride must be the "null" reaction, which couples a fusion reaction at one site with the inverse reaction at another site [5-7]. Such a process is exactly resonant up to the difference in phonon exchange at the two sites. The specific example of this process which we consider to be important for understanding the anomalies in metal deuterides are two-site null reactions of the form

$$\left(d + d\right)_a + \left(^4He\right)_b \leftrightarrow \left(^4He\right)_a + \left(d + d\right)_b$$
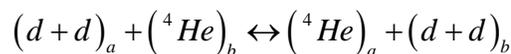
This kind of reaction is illustrated in Figure 3.1. We presume sufficient phonon exchange (with accompanying angular momentum exchange) at both sites to satisfy the microscopic E2 (electric quadrupole) selection rule. This kind of second order site-other-site null reaction is a prediction of the lattice resonating group method outlined above. There has been no consideration of such a process in the nuclear physics literature.

We will argue in what follows that there exists experimental support for the existence of this new kind of reaction process.

$$(d+d)_a \qquad\qquad (d+d)_b$$
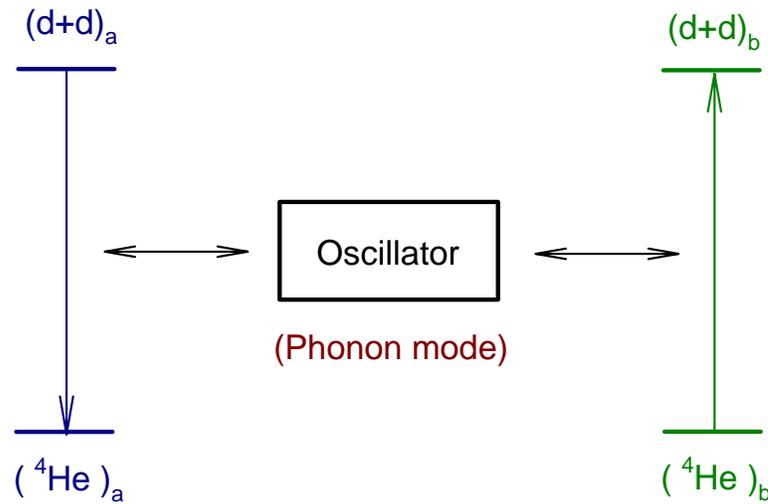
Figure 3.1 A pair of two-level quantum systems coupled through an off-resonant oscillator. Phonon exchange with a common highly excited phonon mode is proposed to allow coupling between nuclear reactions at different sites. An excitation transfer of this type between equivalent systems is termed a "null" reaction.

Many-site null reactions

Within the solid, one expects that there might be many deuteron-deuteron pairs which might be capable of interacting with a given helium nucleus through a second-order off-resonant null reaction process as outlined above. There may also be many helium nuclei which can interact with a given deuteron-deuteron pair as well. We need to extend the model to include interactions that are more general than the simple two-site null reaction process outlined above.

Deuterons in a metal deuteride occupy interstitial sites, such as the tetrahedral or octahedral sites, in preference to substitutional sites. Deuterons at neighboring interstitial sites have a very low probability of coming together sufficiently close to fuse. Our interest is focused instead on molecular D_2 states within the metal (the local two-deuteron state within a single site that is locally spherically symmetric is approximately the same as that of a free molecule of D_2). In PdD, when the deuterium loading exceeds unity, the probability of molecular state [8-10] formation improves. In a metal deuteride with many host metal vacancies, the formation of the molecular state within the metal becomes more probable. The probability that two deuterons can come close enough to fuse is still very small, but it is much higher in the molecular state than for deuterons at neighboring sites. Helium is ubiquitous. Metal deuterides contain small and uncontrolled amounts of helium (primarily ^{4}He).

The size of a phonon mode in a metal deuteride depends on the frequency and wavevector of the phonon mode. The phonon modes in a metal deuteride are very lossy near room temperature, and can be damped in a time on the order of ten oscillation cycles in the case of optical phonons. The size that might be associated with suitable phonon modes are likely to be on the order of hundreds to thousands of

Angstroms in each direction. Within the volume of a phonon mode may exist a very large number of molecular state deuterons, and a few helium atoms.

To extract a useful model out of the multi-site version of the problem, a variety of approximations are required. We can take advantage of infinite-order Brillouin-Wigner theory in order to develop approximations that provide some insight as to the physics involved. The basic picture is that we have many sites, each of which is very nearly a local problem unto itself, with coupling provided by a common phonon mode that interacts with nuclei at the different sites. The Brillouin-Wigner theory allows us to examine how the local molecular D_2 states are impacted by the existence of the second-order site-other-site reactions. The resulting model might be cast in the form

$$E\Psi_{i,n} = \left\langle \Phi_i^a \mid H_a \mid \Phi_i^a \right\rangle \Psi_{i,n} + \Sigma_b \Sigma_{j,n'} \Sigma_{k,n''} \left\langle \Phi_i^a \Phi_i^b \phi_n \mid H - E \mid \Phi_j^a \Phi_j^b \phi_{n'} \mid \frac{1}{E-H} \left\langle \Phi_j^a \Phi_j^b \phi_{n'} \mid (H-E) \mid \Phi_k^a \Phi_k^b \phi_{k,n''} \right\rangle \right\rangle$$

The upshot of this model is that the local molecular deuteron channel at site a ($\Psi_{i,n}$, associated with reaction channel i and n phonons) sees a modified molecular-type potential within the local metal environment $\langle \Phi_i^a \mid H_a \mid \Phi_i^a \rangle$, and has a source which is due to local ^{4}He dissociation.

We have succeeded then in using the lattice-generalization of the resonating group method from the nuclear fusion literature to describe near resonant second-order lattice-induced coupling between molecular-state deuteron pairs in the metal deuteride and helium nuclei. The resulting model constitutes a new kind of solid-state many-body model that describes the interaction of the nuclei and the lattice.

Compact states

The new solid-state many-body problem for the coupled nuclear and lattice system is complicated, and it is not particularly evident at the outset as to what kind of states are predicted by such a model. Based on the calculations that we have carried out so far, it appears that this new many-body model predicts a new kind of compact two-deuteron state. The second-order coupling between the fusion reaction at one site and the dissociation reaction at another site leads to the introduction of a closely spaced deuteron pair at the site where the helium dissociation occurs. Due to the Coulomb repulsion between the nuclei, the two deuterons created from the dissociation process would like to move apart. Unfortunately, this requires that the deuterons tunnel apart, which is a slow process since it is inhibited because the associated barrier is so large.

Such an effect can be seen with the aid of a simple one-dimensional analog model. One often develop seeks simplified versions of a complicated many-body model from which the relevant new physics can be seen and studied in isolation from the difficulties associated with the full theory, so that one can understand things simply. In this case, a convenient analog is constructed by replacing the local molecular state with a one-dimensional potential well. The source term due to ^{4}He dissociation appears as an exchange potential. The relevant one-dimensional analog model can be written as

$$E\psi(x) = \left[-\frac{h^2}{2\mu} \frac{d^2}{dx^2} + V(x) \right] \psi(x) - Kf(x) \int f(y)\psi(y)dy$$

where *V(x)* is the one-dimensional equivalent molecular potential
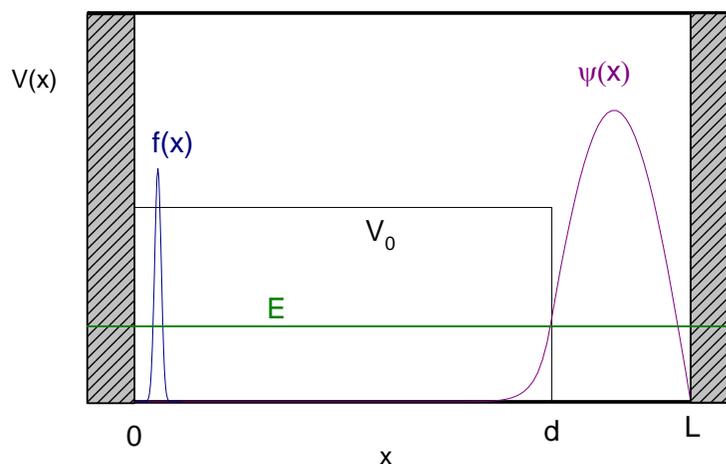
Figure 3.2 Schematic of 1-D analog model. The molecular potential is modeled by a square well with zero potential between d and L, and a constant potential below d. The unperturbed ground state (analog for the molecular ground state) is illustrated as $\psi(x)$. Dissociation of helium leads to two deuterons with a tiny separation. This is accounted for in the function $f(x)$.

We have taken $f(x)$ to be a delta function located near the origin. The strength of the null reactions is modeled in the constant K. This is illustrated in Figure 3.2.

This analog model problem is easily solved. When the coupling constant K is small, the solutions consist of states that are very close to the bound states of the well that contain a small amount of admixture from a localized state near the origin. The associated intuition is that the deuterons spend part of their time in the molecular state, and part of the time localized. We associate the localized component as being due to contributions from deuterons at close range which are produced from helium dissociation, which tunnel apart.

When the coupling constant K is large, then a new compact state forms (see Figure 3.3), with an energy that depends on the coupling strength. The corresponding interpretation is that the deuterons that are created at close range try to tunnel apart, but ultimately come back together to make helium (sending the excitation elsewhere) before they can tunnel apart.

The Kasagi experiment: Observation of compact states

The model outlined above suggests that new compact two-deuteron states may exist in a metal deuteride that has molecular two-dueteron states, helium, and a highly excited THz phonon mode. As the states are expected only to be a few fermi in size, one would have to develop a nuclear diagnostic to detect them, assuming that enough of them were present to give rise to a signal. It may be that such states have already been observed. Specifically, they may have been observed in experiments reported recently by Kasagi and coworkers at Tohoku University in Japan.
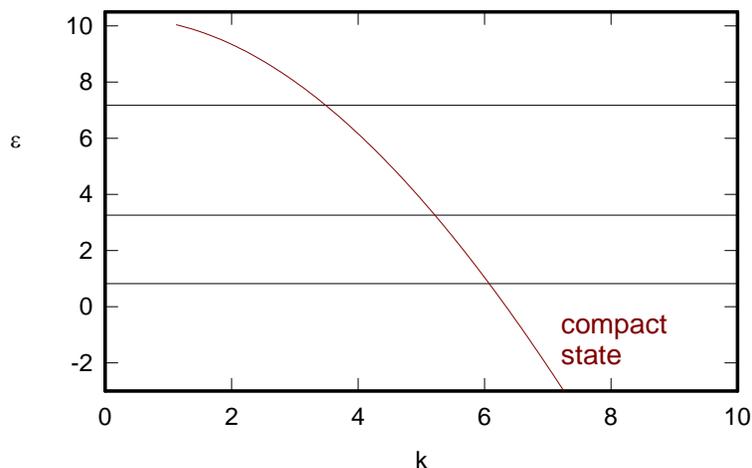
Figure 3.3 Normalized eigenvalues ε as a function of the normalized coupling strength *k* for the square well analog. When the coupling strength increases to a sufficiently large value, a new state appears, with an energy that depends of the strength of the coupling.

In experiments looking at reaction products from 100 keV deuterons on TiD metal deuteride targets, Kasagi observed unexpected proton and alpha particles with a broad energy distribution up to very high energies [11]. These products could not be associated with any conventional fusion reaction. Kasagi noted that the broad energy distribution was consistent with a 3-body exit channel, and that the high energy endpoints were consistent with the reaction

This experiment has been replicated at several laboratories. Last year, this effect was observed in a replication effort at NRL. We conjectured that the Kasagi experiment can be interpreted as detecting the compact dd states discussed above [12], which would provide experimental support for the existence of site-other-site null reactions. A weakness of the Kasagi experiment is that the alpha particles and protons were not measured in coincidence. Consequently, it is possible that the associated signals might somehow arise from some other source. An improved version of the Kasagi experiment to measure the alpha and proton signals in coincidence is planned in the near future.

Absence of Kasagi two-body channels

If compact two deuteron states exist, as suggested by the model, and as indicated by experiment, then one must address the question of stability. Stationary deuterons in close proximity (10-20 fermis) would be expected to fuse on a sub-femtosecond timescale. Yet in the Kasagi experiment, the fraction of all deuterons in compact states is on the order of 10^{-5}, which is inconsistent with such a short lifetime.

Within the framework of the model, there exists the possibility of exchanging many phonons. As remarked upon earlier, phonon exchange has the capability to transfer phononic angular momentum to the microscopic nuclear scale. About 25 units of angular momentum in the compact state is sufficient to stabilize the state against decay by two-body channels, as the associated centripetal potential becomes too great for the exit channel products to tunnel through.

If the compact states are indeed high angular momentum states, then there should be observable consequences. One such consequence is the supression of two-body exit channels in the three-body Kasagi reaction. For example, one might have expected a significant yield for the two-body reactions

$$d + d + d \rightarrow ^4He + d$$

$$d + d + d \rightarrow t + ^3He$$
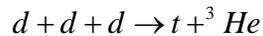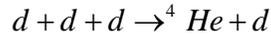
Neither of these reactions are observed in Kasagi's experiments, or at NRL.

We have noted that the absence of these two-body channels can be accounted for through the development of compact states with high angular momentum. For example, a high angular momentum compact state would have an associated large centripetal potential in the two-body exit channels, causing a suppression of these reactions [6,7]. This is the only possible mechanism for stabilizing the ^3He+n channel, since the neutron has no charge.

Proton and deuteron compact states

Although our discussion has focused on compact states formed from two deuterons, there does not seem to be any reason that similar physics should not apply in the case of the proton deuteron system. The associated two-site null reaction is

$$\left(p + d \right)_a + \left(^3He \right)_b \leftrightarrow \left(^3He \right)_a + \left(p + d \right)_b$$
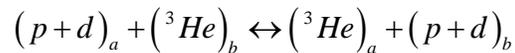
Proton-deuteron compact states would be expected to be present in mixed metal hydride and deuteride system, under assumptions that are analogous for the compact two-deuteron states discussed above. As there exist reports of anomalies in light water systems, we would anticipate that compact proton-deuteron states would be involved, since light water systems have significant (part per thousand) concentrations of deuterium present.

In principle, it should be possible to observe three-body input channel reactions in this system. The analog to the Kasagi reaction in this case is

$$p + d + t \rightarrow ^4He + p + n$$
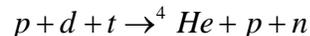
To test this reaction channel would require running a high flux beam of 100 keV tritons on a mixed metal deuteride hydride target. Such an experiment is not being contemplated at present.

An alternate route might be to set up conditions in which the two-body exit channels become allowed. For example, if and energetic alpha particle were incident at 5-10 MeV, then one might hope to observe reactions such as

$$\alpha + d + d \rightarrow ^4He + ^4He$$

$$\alpha + p + d \rightarrow ^4He + ^3He$$
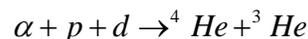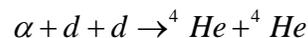
If these reactions were observed, one would be able to infer the amount of local angular momentum associated with a compact state from the dependence of the yield on the incident alpha energy. An exploratory experiment at NRL verified that there are no observable products from proton irradiation at low energy of a metal deuteride that showed the Kasagi effect under equivalent deuteron irradiation [13].

Relation between the Kasagi effect and low-level fusion reactions

If insufficient initial phonon excitation is present, so that the angular momentum transfer to the compact state is in the range of 10-20 angular quanta, then the compact state is unstable against decay by the $p+t$ and $n+^3He$ branches of the dd-fusion process. When this happens, compact state formation would be accompanied by low-level dd-fusion reactions, which would be observable through energetic n, p, t and ^3He emission. This is proposed as a mechanism to account for the Jones effect, which consists of the observation of low-level dd-fusion reactions in metal deuterides. We note that the probability that deuterons tunnel through the Coulomb barrier in a thermal metal deuteride is much too low to lead to fusion reactions at an observable rate.
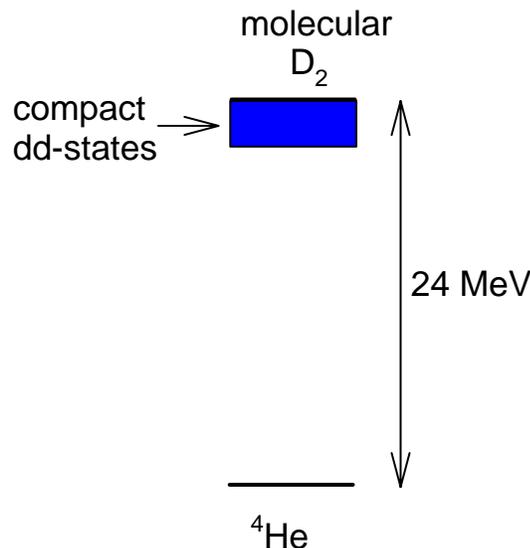

Figure 3.4 Schematic of a "weak'' coupling version of the compact state energy distribution. In this case, compact state formation occurs at energies slightly below the molecular D_2 state energy. In the event that coupling occurs to states with less than 20 units of angular momentum, then conventional dd-fusion reactions would be expected as an allowed decay route for these low angular momentum compact states. An accumulation of compact states with energies near the molecular state could also lead to energy transfer to the host lattice nuclei, giving rise to fast ion emission of the type observed by Chambers and by Cecil.

Relation between the Kasagi effect and experiments of Chambers and of Cecil

The second-order multi-site null reaction process and compact state formation which has been discussed constitutes an example of an interesting but obscure quantum effect in which a large energy quantum is transferred from one (approximately) two-level quantum system to another two-level quantum system through off-resonant coupling with a low energy oscillator. We would expect that a similar process should be able to transfer energy from a two-level system indirectly so as to drive transitions in a continuum. This is illustrated in Figure 3.5.

For example, a 24 MeV quantum transferred to a host metal lattice nucleus would produce very energetic ejecta. In the case of PdD, alpha particles would be ejected with energies in the range of 18 to 21 MeV [6,14]. This is consistent with the observation of Chambers of alpha emission in this range from PdD samples. One would also expect protons, deuterons, tritons and ^3He ejection as well. Such ejecta were observed by Cecil from TiD, with the different products exhibiting energies in the range of what might be expected from the transfer of 24 MeV to the titanium isotopes. Energetic alpha particles from layered thin film metal deuteride targets made of nickel and palladium were recently observed by Lipson [15].
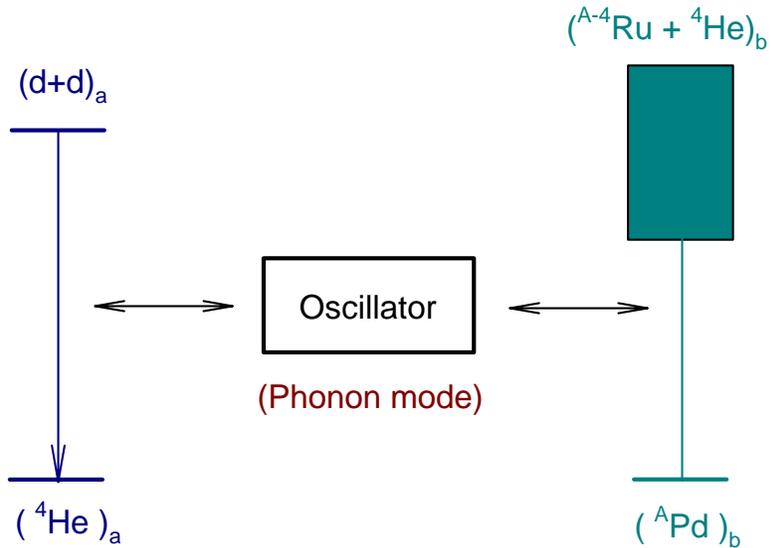
Figure 3.5 Schematic of off-resonant coupling between a two-level system and a transition into a continuum. Compact dd-states with energies near the molecular limit at one site would be capable of an off-resonant coupling to host Pd nuclei at another site that would lead to alpha ejection in the range from 18-21 MeV, as observed by Chambers.

Relation between the Kasagi effect and tritium production

There have been many observations of the development of tritium in metal deuterides (for example see [16]). In the experiments of Claytor at LANL, tritium generation was not accompanied by any detectable neutron emission, either at 2.5 MeV or at 14 MeV, indicating that it is not formed through the tritium branch of the vacuum dd-fusion reaction, and that the tritium has an energy of less than about 10 keV when first formed.
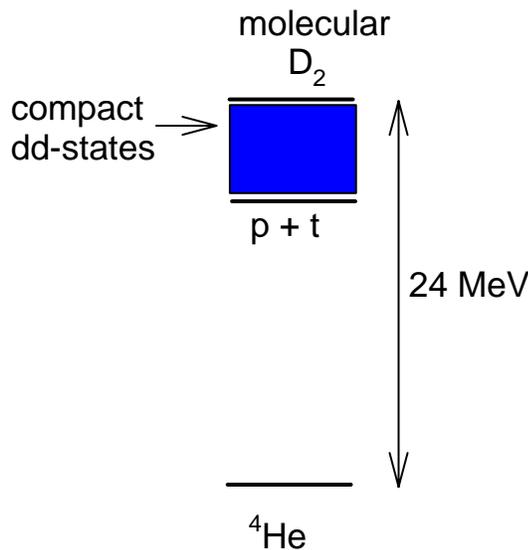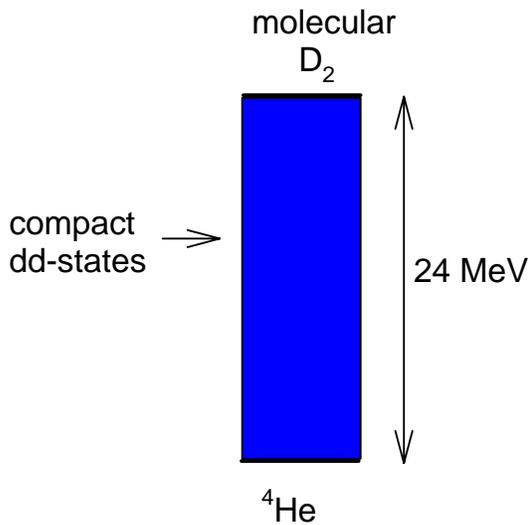
We are considering that the compact deuteron-deuteron states may be responsible for this effect [6]. For example, the model described above suggests that the compact states can occur at different energies, dependent on the strength of the coupling constant. If there develops an effective continuum of compact states with energies uniformly distributed between the molecular state energy down about 4 MeV to the $p+t$ state energy, then one can envision a ladder process in which the population of compact dd-states exchanges energy with the phonon field, tens of quanta at a time, in a very large series of sequential exchanges (all of these mediated by second-order two-site null reactions). This is illustrated in Figure 3.6.

Figure 3.6 Schematic of compact state energy distribution consistent with tritium production.

Relation between the Kasagi effect and the Pons-Fleischmann effect

The basic Pons-Fleischmann effect is the development of excess heat from a metal deuteride. Observations of this effect have by now been reported from many laboratories. Bush, Miles and coworkers reported the first observation of a correlation of [4]He with the excess heat [17]. Other laboratories have reported a quantitative correlation between the excess energy and the amount of helium produced. The most accurate of these experiments indicates a reaction Q-value of 24 MeV to within 10-15%.

The connection between this effect and the model under discussion lies within the strong coupling limit of the model, in which a continuum of compact states is produced between the molecular state energy and the [4]He state energy [6]. The conversion of the nuclear energy to phonon energy occurs freely between the compact states and the lattice. Following a very large number of sequential transitions, the available reaction energy is extracted from the compact states, leaving [4]He as an ash. This is illustrated in Figure 3.7. This basic mechanism is in agreement with the experimental observations.

molecular
D_2

compact
dd-states

24 MeV

[4]He

Figure 3.7 Schematic of the compact state distribution in the strong coupling limit. In this case, compact states can exist with energies between the molecular state energy and the [4]He energy. This allows for continuous exchange of nuclear energy between the deuterons and the lattice.

Let us step back for a moment, and try to form a high level picture as to what is going on in the excess heat experiments from an engineering point of view. Suppose that we wished to design new systems that make heat along the lines outlined above. To do so, we wish to develop molecular state deuterium inside the metal deuteride along with [4]He as the initial condition. Molecular state formation requires either very high loading, high temperature, or lots of vacancies (or combinations of these). Once the system is properly prepared, then we wish to impose a very strong high frequency phonon field. Optical phonons or THz-level acoustic phonons are believed to be most effective in this process (there is not yet experimental support for this). Alternatively, there is strong mixing between the phonons and electronic states, such that excitation at higher energies may lead to a plasmon response with a phononic admixture. The phonon coupling mediates transitions from the molecular states to the compact states, as well as the exchange of nuclear energy with phononic energy among the near-continuum of compact states. The associated density of states is potentially an observable in the Kasagi experiment [18]. If the process is successfully executed, then deuterium is burned to make [4]He. At least, this is what we would deduce based on the model and based on the experimental data set as we presently understand it.

Discussion

We have developed a new picture and associated model for the anomalies in metal deuterides. The anomalies include a variety of effects, some of which we have discussed briefly here (Kasagi effect, low-level fusion reactions, alpha emission, tritium production and heat and helium generation), and some of which we have not (the Wolf effect, which involves the activation of host lattice metal nuclei, and the recent results of Swartz, who observed an enhancement of excess heat in a light water system with the addition of heavy water [19]).

Over the past several years, a new model has evolved that addresses these anomalies on a unified footing. The premise of the new model is that solid-state effects need to be included intimately at the outset. The coupling between nuclei and phonons leads to the exchange of angular momentum between the phonons and the microscopic nuclear system, and results in the possibility of new second-order site-other-site reactions. A fusion reaction at one site can be coupled to a dissociation reaction at another site. The many-body version of this kind of reaction leads to the prediction of new compact two-deuteron states. In essence, the many different anomalies that have been reported can be seen simply as a consequence of the presence of these new compact states. Moreover, the results of the Kasagi experiment are consistent with the direct observation of compact two-deuteron states. The model predicts that these states can have a broad energy distribution, which is consistent with the observation of tritium generation. and with heat associated with quantitative helium generation

We conclude that order is beginning to emerge from the chaos of the field, and that many experimental observations which previously seemed to make little sense can now be seen as being closely related to one another. The evidence supports the existence of a new underlying physical effect, which is very exciting. The possibility of designing and engineering new devices based on the new physics described here is not only exciting, but has the potential to open the door to new technologies which could have a major impact on society.

[1] J. A. Wheeler, *Phys. Rev.* **52**, 1107 (1937).

[2] A. M. Lane and D. Robson, *Phys. Rev* **151**, 774 (1966). D. Robson and A. M. Lane, *Phys. Rev.* **61**, 982 (1967).   A. M. Lane and D. Robson, *Phys. Rev.* **185**, 1403 (1969). R. J. Philpott and J. George, *Nucl. Phys.* **A233**,164 (1974).

[3] P. L. Hagelstein, *Phil. Mag. B* **79**, 149 (1999).

[4] T. Hamada and I. D. Johnston, *Nucl. Phys.* **34** 382 (1962).

[5] P. L. Hagelstein, "Anomalous Energy Transfer,'' *Proceedings of the Seventh International Conference on Cold Fusion*, Vancouver, March 1998.

[6]   P. L. Hagelstein, "A unified model for anomalies in metal deuterides,'' *Proceedings of the 8th International Conference on Cold Fusion*, Lerici (La Spezia), Italy, May 2000; page 363.

[7] P. L. Hagelstein, "Theory for Anomalies in Metal Deuterides,'' *Trans. Am. Nucl. Soc.* **83**, 359 (2000).

[8]   K. P. Sinha and Peter L. Hagelstein, "Electron screening in metal deuterides,*'' Proceedings of the 8th International Conference on Cold Fusion*, Lerici (La Spezia), Italy, May 2000; page 369.

[9] K. P. Sinha and P. L. Hagelstein, "Electron screening in metal deuterides,'' *Trans. Am. Nucl. Soc.* **83**, 359, (2000).

[10]   K. P. Sinha and P. L. Hagelstein, "Electron screening in metal deuterides,'' *Bull. APS*, **45**, 235 (2000).

[11] J. Kasagi, T. Ohtsuki, K. Ishu and M. Hiraga, *J. Phys. Soc. Japan* **64**, 777 (1995).

[12]   P. L. Hagelstein, "A proposed explanation for the Kasagi effect,'' *Bull. APS* 2001.

[13] G. Hubler, unpublished.

[14] P. L. Hagelstein, "A model for fast ion emission in metal deuterides," *Bull. APS* **45**, 235 (2000).

[15] A. G. Lipson et al, "In-situ long range alpha particles and x-ray detection for thin-film Pd cathodes during electrolysis in Li2SO3/H2O," *Bull. APS* **47**, 1219 (2002).

[16] M. McKubre, F. Tanzella, P. Tripodi and P. L. Hagelstein,"The emergence of a coherent explanation for anomalies observed in the D/Pd and H/Pd systems: Evidence for [4]He and ^{3}H production," *Proceedings of the 8th International Conference on Cold Fusion*, Lerici (La Spezia), Italy, May 2000; page 3.

[17] B. E. Bush, J. J. Lagowski, M. H. Miles, and G. S. Ostrom, "Helium production during the electrolysis of D_2O in cold fusion experiments," *J. Electroan. Chem.* **304**, 271 (1991).

[18] P. L. Hagelstein, "Extracting compact dd state energies from the Kasagi experiment," *Bull. APS* **47**, 1219 (2002).

[19] M. Swartz, "The impact of heavy water (D_2O) doping," *Bull. APS* **47**, 1219 (2002).