Energy-related Problems

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Introduction

Our focus in recent years has been on applied problems relating to an unconventional approach to energy generation, and additionally focused on the general problem of thermal to electric energy conversion. At present, the energy problem continues to haunt our nation and the world in general. Gasoline prices have been high in various parts of the US during the last year, in part due to uncertainties associated with possible military action in the Middle East. The US presently has troops in Iraq, which is perceived in different parts of the world as being due to an interest in the natural oil reserves within that country. The world very much needs solutions to the energy problem, as energy issues have become of growing importance as time goes on.

In this report, we discuss results that we have obtained in the three different areas that we have focused on recently.

In the first article, we discuss recent advances in the understanding of the thermal diode for thermal to electric energy conversion. We have reported previous on our theoretical approach and on experiments on thermal diode devices. The basic idea initially was to implement a solid-state version of the vacuum thermionic converter using semiconductor technology. A highly doped hot region would serve as an emitter. Electrons would ``boil'' out of the emitter, and be collected at a cold contact. The voltage induced in this approach could be used to drive an electrical load. The early experiments that we performed indicated that the approach was a very good one, and the efficiencies that were observed were higher than what could be accounted for theoretically with the models that we were studying at the time. A lack of understanding of precisely how the devices work has led to a lack of acceptance on the part of the relevant scientific community, and we have been motivated to clarify the relevant mechanisms. Recently, some significant progress has been made in this area. We proposed that a thin p-type layer between the emitter and solid gap could isolate the emitter region from the solid gap region, and thereby allow a discontinuity in the fermi level to arise. Recent experiments appear to confirm this idea, and devices constructed with a controlled p-type separation layer have shown improved performance systematically. Results from a simple nonperturbative transport model appears to support to conjecture that the isolation of the emitter can lead to an elevated fermi level at the edge of the solid gap region. The current and voltage results can be thought of in light of these

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results as a device composed of two thermoelectric materials, where the solid gap region acts as a conventional thermoelectric and the very thin junction region acts as a material with an anomalously high thermopower. The thin (micron) junction region in this case dominates the thermally induced voltage of the thick (millimeter) solid gap region. This is an important result, as it points to the possibility of developing a new class of solid-state converters with near-Carnot limited thermal to electric conversion efficiencies.

In the second article, we review progress that we have made on the problem of anomalies in metal deuterides. Research in this area is controversial, as the scientific community at present does not believe that there are any anomalies in metal deuterides, while a few hundred scientists from around the world who have studied them are equally convinced that they are very real. As year after year goes by, there seems to be no hope of resolving things. The detectors appear to see neutrons, charged particles, tritium, excess heat and other anomalies. Papers submitted to mainstream journals do not fare well as referees point out that all such effects are impossible according to ``conventional'' nuclear physics. The possibility that new physics might be involved is not seriously contemplated. We recently put together a conference proceeding covering results from the past couple of years, and a final report that included results more generally from over the past half-decade or more. In both cases, it was apparent that a great many results have been obtained, and that there was no article that provided an overview of the associated picture. We have taken material from the executive summary of the final report to present here in the second article. The basic approach is to view the anomalies ultimately as a consequence of phonon-exchange in nuclear reactions. Phonon exchange on the microscopic level should allow for selection rules to be satisfied in a fusion reaction without gamma emission. Reactions occurring at different sites can be coupled into a new kind of second-order process should both exchange phonons with a highly excited phonon mode. Experimental results on anomalies in metal deuterides give results that appear to be consistent with such a picture. A many-site version of the basic model, in which all reactions that exchange phonons with a common highly excited phonon mode are coupled together, appears to lead to an interesting unified model which predicts low-level fusion in one regime, fast ion emission in another, an anomalous three-body reaction as claimed to have been seen by Kasagi and by others, an excess heat effect, and tritium production – all of which are now claimed to be observables in experiments on anomalies in metal deuterides.

In the third article, we describe a new approach to the problem of thermal to electric energy conversion based in part on a second-order quantum excitation transfer effect that we have discussed previously in these reports, and in part on a novel associated conversion scheme. In our initial proposals, we worked with excitation to interband transitions in the hope of making use of conventional photodetector technology, much as is done in the case of thermophotovoltaics. However, the coupling in the case of an interband transition is relatively weak in comparison to the coupling available from intersubband transitions. Unfortunately, there does not appear to be an equivalent photodetector technology that can be adapted to make use of such an approach. Recently, we noticed that there exists a class of single carrier energy conversion schemes that should be possible that might be able to convert excitation produced by the second-order quantum effect into electricity. In the third article, we describe this general approach briefly.

1 Thermal to Electric Conversion with Thermal Diodes

Project Staff
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Several years ago, we were interested in the problem of the possible extension of the vacuum thermionic scheme to the solid state under conditions where transport in the solid gap region occurred by diffusion. In such a scheme, hot electrons would boil out of a solid emitter region at high temperature, transport diffusively across a solid gap region, and then be collected at a cold metal contact. Devices of this type were fabricated in InSb and HgCdTe implementations, and were found to give conversion efficiencies that were significantly enhanced over what might be expected from equivalent bulk thermoelectrics.

It was soon recognized that these devices operated far better than would have been expected based on the models then under consideration. Moreover, the devices appeared perhaps to operate in violation of the Onsager current relation

$$\mathbf{J} = \sigma\left(\frac{1}{q}\nabla\varepsilon_{\mathrm{F}} - \Sigma\,\nabla\mathrm{T}\right)$$

We made a number of attempts at modeling the new devices over the years with varying degrees of success. For example, it was found that the experimental data could be adequately modeled through an empirical model for the solid gap region of the form

$$\mathbf{J} = \sigma\left(\frac{1}{q}\nabla\varepsilon_{\mathrm{F}} - \Sigma\,\nabla\mathrm{T}\right) + \mathbf{J}_{inj}$$

where $\mathbf{J}_{\mathrm{inj}}$ represented an injected current density from the emitter.

To predict the injected current density, we studied various abrupt junction models in which the forward current from the hot emitter region and reverse current from the solid gap region were analyzed. We discovered a new second-order thermionic current injection effect in which the forward and reverse currents do not balance across a junction with no temperature discontinuity, as long as the individual electron distributions have significant components that differ from the Fermi-Dirac distribution. Such models appeared to predict current injection at the level observed in the experiments when adopted as a boundary condition.

Unfortunately, efforts to extend such models to the more general case of a finite junction, requiring self-consistency with the transport equation and Gauss's law, ultimately failed. The reason for this is that it is impossible to support a significant discontinuity in the Fermi level across an *n*n* junction, as the associated high conductivity shorts it out. We were not able to develop more than O(100 μV) of voltage across an *n*n* junction, taking account of the second-order thermionic injection mechanism outlined above. The experimental results indicate the presence of up to a O(50 mV) voltage discontinuity in the thermal diodes tested.

Separation of emitter and gap regions with a *p*-type layer

We presented a possible solution to this problem at the International Thermoelectrics Conference ICT02 last August. If a voltage discontinuity is shorted out across an *n*n* junction, then the problem should be fixed if there were a *p*-type region separating the *n**-type emitter region and *n*-

type solid gap region. The proposal was to examine transport an *n*pn* model for the thermal diode, under the assumption that a thin *p*-type region was present in the initial sets of experiments of which that we were not. During the fall of 2002, experimental evidence in support of the presence of a *p*-type region near the emitter was developed when thermal diodes that worked well were analyzed. This provided the impetus to build new devices in which a thin controlled *p*-type layer was implemented as part of the device design, and the acceptor doping level was varied. The doping profile is shown in Figure 1.1, and a summary of the results is given in Figure 1.2.

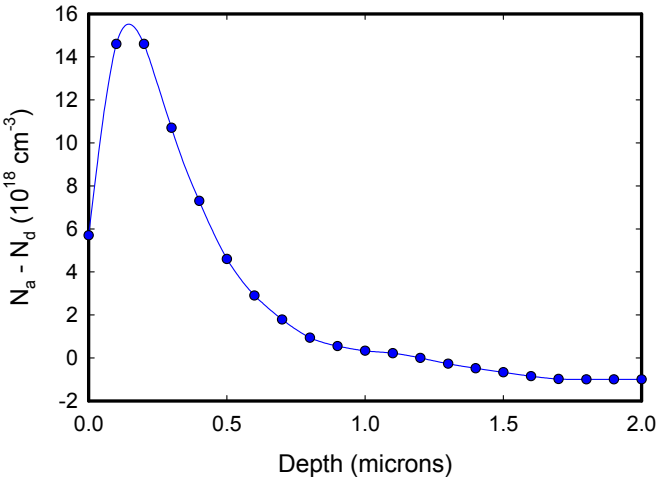

Fig.1.1: Shown here is the acceptor (defect) concentration minus donor (Te) concentration in InSb thermal diode following ^{4}He implantation at six beam energies between 20 keV and 350 keV. The donor doping in the solid gap region is 10^{18} cm^{-3}.

The recent experiments support the conjecture that a p-type region is required to support a discontinuity in the Fermi level between the emitter and solid gap region in the thermal diode. These observations have led to a significant improvement in our ability to implement emitter-layers in thermal diodes that produce an enhancement in thermal to electric conversion efficiency over the thermoelectric limit.

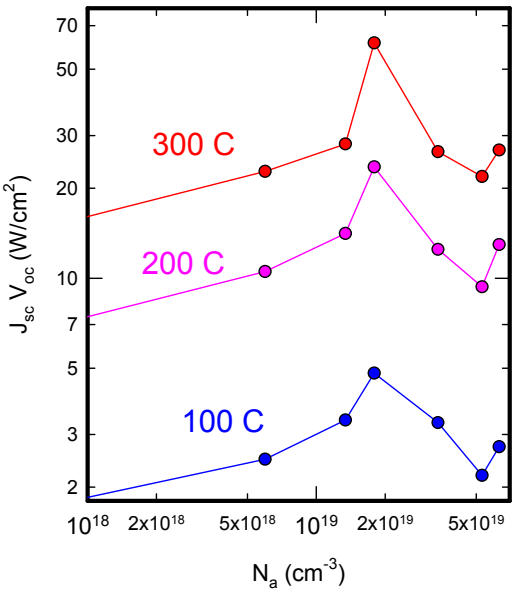

Fig. 1.2: Here we plot the product of short circuit current density and open circuit voltage for InSb thermal diodes as a function of acceptor concentration. The emitter in this case is a metal contact, and the p-type layer is implemented by helium bombardment as shown above, with different defect concentration densities. The thermal diodes tested here were 1 mm thick. We see that there appears to be an optimum in the acceptor concentration near $1.8x10^{19}$ cm^{-3}. The peak enhancement in the $J_{sc}V_{oc}$ product over thermoelectric performance is about a factor of 6.

Transport Modeling

As indicated above, it has not proven easy to develop an adequate theoretical model for these results based on previous experience with thermoelectric semiconductors and junctions in the literature. At ICT02, we outlined a new approach that we had begun to explore to address this kind of problem. In this approach, we begin with a steady-state transport equation in the relaxation time approximation

$$\left[\frac{1}{\hbar}\nabla_{\mathbf{k}}\varepsilon\right] \cdot \nabla f - \left[\frac{1}{\hbar}\nabla\varepsilon\right] \cdot \nabla_{\mathbf{k}} f = \frac{f_0 - f}{\tau_s}$$

We have found previously that the thermoelectric performance of InSb and HgCdTe are adequately described using an energy-independent (but density-dependent) relaxation time approximation. It is possible to solve this transport equation formally using an integral solution, similar to what is done for radiation transport problems. If we transform to a coordinate system for the one-dimensional version of the problem in which the electron energy ε is taken to be one coordinate, and the length along a trajectory s is adopted for another coordinate, then the transport equation can be written in the same form as for radiation transport problems

$$\frac{df}{ds} + \kappa f = S$$

A formal solution to the transport equation can be developed that is similar in form to the case of radiation transport problems

$$f = \int e^{-\tau}\left[S_{-}(\tau)\Theta(k_z) + S_{+}(\tau)\Theta(-k_z)\right]d\tau$$

In this case τ is the associated ``optical depth'' for scattering along a curved trajectory. The source function in this model is the Fermi-Dirac function

$$f_0 = \frac{1}{e^{(\varepsilon-\varepsilon_F)/k_B T} + 1}$$

The current density in this approximation is then given formally by

$$J = -q\left\langle \mathrm{v}(\mathbf{k}) \int d\tau e^{-\tau}\left[\left(\frac{1}{e^{\frac{\varepsilon-\varepsilon_F}{kT}}+1}\right)_{-}\Theta(k_z) + \left(\frac{1}{e^{\frac{\varepsilon-\varepsilon_F}{kT}}+1}\right)_{+}\Theta(-k_z)\right]\right\rangle$$

An interesting feature of this formulation is that the temperature and fermi level are now associated with the source function rather than with the distribution function. If no gradients occur on the spatial scale of a scattering length or smaller, then the distribution approaches the Fermi-Dirac distribution with linear corrections due to conductivity and thermopower. However, in the presence of sharp gradients on the scale of a scattering length, the temperature and fermi level are well defined in terms of the source function under conditions where the distribution function may be very different from a Fermi-Dirac distribution.

A key feature of the thermal diode is that an enhancement in the open circuit voltage is observed at zero current in the presence of a temperature gradient. This is very difficult to understand in light of the Onsager current relation in the absence of an empirical injection current in the bulk region. If one demands a completely self-consistent model, then the contribution of the solid gap region must be precisely the same under zero current conditions as an equivalent bulk thermoelectric. Consequently, all of the voltage enhancement under zero current conditions must be associated with the emitter structure. Hence a calculation of the fermi level in the vicinity of the emitter and the edge of the solid gap region under zero current conditions should show an anomalous increase in the fermi level in light of the experimental results.

To investigate this, we have developed a computer code that determines the source function fermi level in the presence of a linear temperature gradient in the vicinity of an emitter structure with a *p*-type layer separating the emitter from the solid gap. We have assumed a symmetric model doping profile consistent with an electrostatic potential of the form

$$\psi = \psi_0 + \Delta\psi_0 \; e^{-\beta z^4}$$

We have found that a model supergaussian potential allows for a simpler interpretation of the results than other model potentials that we investigated, due in part to the absence of a second derivative at the origin.

The calculations appear to show a discontinuity in the fermi level in association with the doping profile, as long as the spatial scale is sufficiently small to give significant changes in the electron density over a scattering length, and also as long as the doping profile includes separation of the *n*-type emitter and solid gap regions with a strongly *p*-type region. Under these conditions, we see a clear enhancement of the source function fermi level. An example of this is shown in Fig. 1.3 in the case of an InSb model.

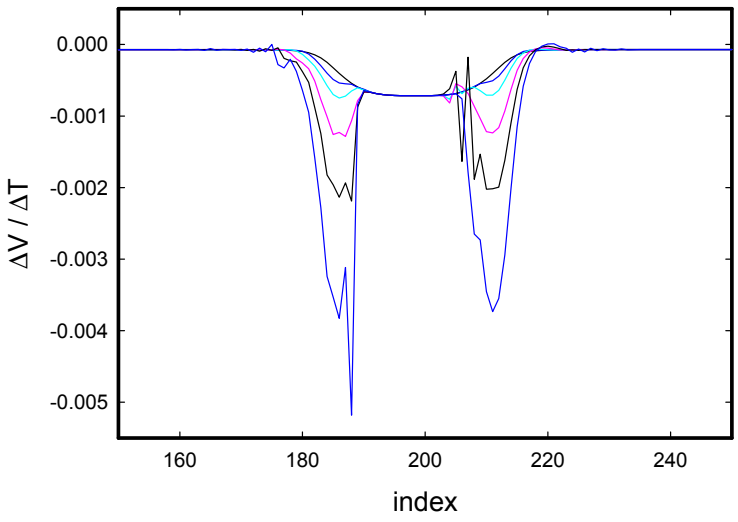

Fig. 1.3: Results for the change in fermi level between neighboring grid points divided by the temperature difference as a function of the spatial grid index. The different curves are for calculations that are identical except for the spatial scale of the doping profile. For a spatial scale more than 10 microns, the ratio approaches the thermoelectric limit of the thermopower. At smaller spatial scales, large increases are observed.

The situation can be seen in the case of results shown in Fig. 1.3, where we have indicated the associated discontinuity in source function fermi level as

$$\frac{\Delta V}{\Delta T} = \frac{1}{\mathrm{q}} \; \frac{\varepsilon_{\mathrm{F}}[i+1] - \varepsilon_F[i]}{T[i+1] - T[i]}$$

There are some oscillations in some of the result due to various technical details associated with the gridding and convergence scheme (recall that the current in this model is strongly nonlocal in the source function fermi level), but the total change in the fermi level does not appear to be particularly sensitive to the effect.

We see that transport effects in this case lead to results that are equivalent to what might be expected if material with a very high thermopower were present. It is well known that the thermopower increases significantly for electrons in a *p*-type region, and this effect can be seen in Fig 1.3. It is also known that the thermopower can be modified by the presence of a gradient in ψ, however, these effects are understood as not changing fundamentally the basic physics of the Onsager current relationship. In our results, we see that the electron Onsager current relation is a consequence of minor perturbations of the electron distribution, which appears to break down when there are significant changes in the potential over a scattering length. In the present case, we are no longer in a region adequately described by a perturbative formulation, and a stronger method is required to analyze the problem.

The results can be understood qualitatively in terms similar to that we have used to discuss the problem previously. Electron injection from the high electron density region into lower electron density region is unbalanced because part of the distribution in both regions deviate from the Fermi-Dirac distribution. We have termed this effect a second-order thermionic injection effect. To counter this under zero current conditions, we require an ohmic return current. If the electron conductivity is restricted due to the relative absence of electrons, then the system responds by increasing the fermi level as a way to generate sufficient ohmic return current to counter the forward injection. The result is a discontinuity in the fermi level, which we have plotted in a way that brings out the difference in the effect from thermoelectric behavior. The situation interestingly enough is similar on in the case of the second junction where the electron density increases. In this case, a relative absence of electrons is injected from the high electron density region backward into the *p*-type region. The system responds once again by changing the fermi level, however, in this case it changes so as to increase the current injection from the high electron density region.

Discussion

We have seen an enhancement of up to 300 in the $\Delta V / \Delta T$ ratio over the thermopower of the solid gap region in calculations so far, limited at this point by technical issues associated with the calculation rather than by physical effects. These results suggest a new interpretation for the experimental results that we have obtained previously. Instead of an anomalous current injection effect, we should think of the thermal diode in regard to the current and voltage relation as simply two thermoelectric materials in series. The solid gap region acts like a normal semiconductor thermoelectric. The thin emitter region, in light of the present discussion, acts as if it as a greatly enhanced thermopower (again, only in regard to current flow). The total open circuit voltage for the two regions is composed of contributions from each

$$\Delta V = \Sigma_{gap}\ \Delta T_{gap} + \Sigma_{eff} \Delta T_{junction}$$

The gap thermopower is on the order of –100 microvolt/K, commensurate with near-intrinsic semiconductor thermoelectric behavior. Most of the temperature drop is across the gap, as we presume that the associated temperature drop is in proportion to the thickness (the gap thickness is on the order of 1 mm). A small amount of temperature drop occurs across the junction (which is on the order of a micron). For the contribution to the voltage of the junction to be commensurate with the contribution of the solid gap region, the effective thermopower must be on the order of 1000 times greater. This is consistent generally with the results we have obtained

from the new transport models. The conclusion from this line of argument is that the thermal to electric performance of very thin junction regions in the vicinity of the emitter is dominating the current and voltage characteristic of the bulk solid gap region. In a sense, this could be thought of as a super-thermoelectric effect.

These arguments suggest that it may be possible to develop solid state thermal to electric converters with even better performance by maximizing the fraction of the device that is junction over solid gap region. For example, one can imagine constructing a semiconductor with a sinusoidal doping profile, in which the number of junctions is on the order of hundreds or thousands. If the interpretation of the thermal diode experiments as given here is correct, then such a device should have an effective figure of merit ZT that is very much greater than unity with an associated near-Carnot limit internal conversion efficiency.

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2 Anomalies in Metal Deuterides

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As late as 2002, the scientific community had not yet appreciated the significance of a variety of experimental results that might be included under the general heading of anomalies in metal deuterides. The prevailing view among nuclear and solid state physicists was that fusion reactions could be adequately described using a formulation that is based on the presumption that reactions in solids work the same way as reactions in vacuum, save for minor issues such as screening effects that can be incorporated as a trivial modification of the basic vacuum result. Referees for mainstream journals continued to reject experimental results that do not conform to this picture, relying in part on the argument that the experimental results are not in agreement with accepted vacuum physics. The possibility that the accepted picture based on vacuum physics might be incomplete could not yet be contemplated within the mainstream. Theoretical work in the area was still considered to be unmotivated due to the relative absence of experimental results in the mainstream literature.

From our perspective, the new experimental results pointed to a different picture. Fast alpha emission around 20 MeV from the Chambers experiment provided evidence of a new kind of site-other-site reaction, in which the reaction energy of a d+d → ^{4}He transition at one site is expressed in alpha ejection from Pd nuclei at another site. Such an effect could only be due to a new second-order process in which both reactions involve phonon exchange with a common highly excited phonon mode. This provides the motivation for developing a new theoretical formulation for nuclear reactions in the lattice, where the lattice is included as an integral part of the problem at the outset. Phonon exchange comes about naturally in such a formulation, and provides some understanding as to what is going on more globally. The dominant site-other-site process in such a picture is a null reaction where a d+d → ^{4}He transition at one site is coupled to the inverse process at another site. A more detailed consideration of such processes suggests that nuclei produced by the dissociation of ^{4}He should have difficulty tunneling apart due to the Coulomb and centripetal barriers, and that a new kind of localized two-nucleus state should occur when a subsequent null reaction occurs before the nuclei can tunnel apart. Some further modeling indicates that the localized states can involve two deuterons, p+t, or n+^{3}He states, the later of which have a greater possibility of occurring at energies near the molecular D_2 state since the nuclear energy difference can offset the localization energy under conditions where the exchange energy due to the null reactions is relatively small. The existence of such localized states is supported by results from the Kasagi experiment, in which an incident deuteron appears to reaction with two deuterons to give a three-body n+p+α final state. Such localized states would require considerable angular momentum for the suppression of the conventional dd-fusion channels. Evidence in support of this comes from the absence of the d+α and t+^{3}He two-body exit channels in the Kasagi experiment, channels that should be suppressed by centripetal barrier effects if significant angular momentum is present initially. The presence of localized states has a dramatic impact on the tunneling probability between deuterons in the molecular D_2 state within the metal deuteride, as the boundary condition near the origin is modified. This finds experimental support in the results of the Jones experiment, in which low-level dd-fusion products are observed under conditions where none are expected based on estimates for the tunneling probability in molecular D_2. The coupling between the nuclear and phononic degrees of freedom when many two-nuclei compact states interact with a strongly excited phonon mode is such that efficient energy exchange between the two degrees of freedom is predicted under conditions where the localized states are stabilized by sufficient angular momentum exchange and are nearly resonant with molecular D_2 states by sufficient null reaction exchange energy. Support for

this comes from experiments in which excess energy is produced in metal deuterides, and ^{4}He is observed in amounts commensurate with an overall a d+d → ^{4}He reaction mechanism.

Within such a picture, the experiments of Pons and Fleischmann are seen as being indicative of a new kind of reaction in which phonon exchange plays an important role. The Jones experiment is interpreted as supporting the existence of new localized states that are nearly resonant with the D_2 state within the metal under conditions where the localized states do not have sufficient angular momentum to be stabilized.

Whether this new picture is correct or not ultimately depends on the results of experiment. The eventual acceptance of this or any other new viewpoint by the scientific community depends less on what happens in the laboratory, but instead more on social issues. In what follows, we outline some of the key results that we have obtained in the course of our theoretical work in the area.

Tunneling in PdD

Early on in 1989 when the Jones effect was first under discussion, there were many manuscripts put forth that discussed the problem of double site occupation in TiD and PdD. The basic issue involved is that the tunneling probability and associated fusion rate for molecular D_2 had been explored, with a very low result for both quantities. As tunneling in TiD was expected to be about the same as for the D_2 molecule, it appeared that the Jones effect could be ruled out based on such theoretical considerations. Subsequent measurements of dd-fusion cross section for low energy (keV) deuterons incident on TiD targets gave deviations from the free space fusion cross section for bare ions that were consistent with screening at a level commensurate with the molecular D_2 problem.

There was further discussion in 1989 that deuterons occupied primarily octahedral sites in PdD and tetrahedral sites in TiD, and that these deuterons were on average further apart than in molecular D_2, and hence would have a smaller associated tunneling probability. These questions were of interest to us over the years, as many speculative papers appeared suggesting that the physics might be otherwise. In addition, when we began focusing on schemes based on dd-fusion reactions, these questions began to become important for our work.

We were interested in the basic question as to what conditions give rise to the largest tunneling rate in PdD. The basic issue in question is that to achieve tunneling at the molecular D_2 level, it would seem that a molecular version of the D_2 molecule would need to be present within the metal deuteride. In the case of double occupancy of a site, perhaps the associated D_2 wavefunction could be approximated by a molecular wavefunction, modified in some way to account for the potential of the surrounding host lattice atoms. Given that the probability for double occupancy in bulk PdD is very low, the associated question arose as to what is the tunneling probability associated with deuterons in neighboring sites.

In response to this, we developed two-deuteron variational wavefunctions for the problem

$$E\Psi\left(\boldsymbol{r}_1,\boldsymbol{r}_2\right)=\left[-\frac{\hbar^2\nabla_1^2}{2M}-\frac{\hbar^2\nabla_2^2}{2M}+V_{mol}\left(\boldsymbol{r}_2-\boldsymbol{r}_1\right)+V_{lat}\left(\boldsymbol{r}_1\right)+V_{lat}\left(\boldsymbol{r}_2\right)\right]\Psi\left(\boldsymbol{r}_1,\boldsymbol{r}_2\right)$$

We studied the problem using wavefunctions of the general form

$$\Psi\left(\boldsymbol{r}_1,\boldsymbol{r}_2\right)=\phi_a\left(\boldsymbol{r}_1\right)\phi_b\left(\boldsymbol{r}_2\right)g\left(\boldsymbol{r}_2-\boldsymbol{r}_1\right)$$

As perhaps might have been anticipated, we found that the tunneling probability associated with deuterons at neighboring sites was astronomically low. The potential barrier associated with realistic potential models is sufficiently high and wide that it introduced tens of orders of magnitude reduction in the tunneling rate over that of the molecular problem. This was true for O-O, O-T, and T-T occupation. We considered separately the cases in which a deuteron at one site tunneled to a neighboring site, and where deuterons from both site tunneled in order to meet in the region between sites.

The basic conclusion is that any reactions involving two deuterons in metal deuterides must involve the molecular D_2 state within the metal. A retrospective analysis of the different conditions under which anomalies have been reported suggests that in all cases the highest level of anomalies are reported in metal deuterides in which the molecular D_2 content is maximized. For example, in electrochemical experiments at SRI, the loading is maximized such that the deuterium concentration exceeds the Pd density near the surface – conditions that would maximize double occupation of a site. Double occupancy is also maximized in the presence of host metal lattice vacancies, and many successful experiments have been reported in materials that would be expected to have very high defect densities. In some cases, experiments operate at elevated temperature with relatively low loading, with positive results. In such cases, the elevated temperature combined with lattices containing large concentrations of defects would maximize double site occupation. We note in addition that host metal lattice vacancies are thermodynamically favored in highly loaded PdD and NiD (Fukai used this feature to create metal hydrides with one out of four host metal lattice atoms missing), such that they will diffuse inward from surfaces at slow rates. We conjectured that this mechanism might have been responsible for a long time constant associated with the excess heat effect in the early SRI experiments.

Dielectric Response at Short Range

Ichimaru published in *Reviews of Modern Physics* a computation of screening between deuterons in PdD and TiD based on relatively sophisticated models that are used in astrophysics. Based on his calculations, he concluded that the tunneling probability is increased by on the order of 50 orders of magnitude from the results of the molecular problem. If true, this would be a very important contribution, and might help to shed light on the problem of anomalies in metal deuterides generally.

In Ichimaru's model, the effect that contributes the largest amount to the screening is a model for the static dielectric constant used within the effective Coulomb interaction. We were unfamiliar with the use of a dielectric response other than the vacuum dielectric response in the case of deuterons close together within the lattice.

To investigate this, we developed a version of a linear response model for the electrostatic interaction between two deuterons in metal deuterides. The result can be expressed in the form

$$\hat{H} = \frac{\left|\boldsymbol{P}_1\right|^2}{2M_1} + \frac{\left|\boldsymbol{P}_2\right|^2}{2M_2} + \frac{q_1 q_2}{\left|\boldsymbol{R}_1 - \boldsymbol{R}_2\right|} + E_e\left(\left|\boldsymbol{R}_1 - \boldsymbol{R}_2\right|\right) + V_{lat}\left(\boldsymbol{R}_1\right) + V_{lat}\left(\boldsymbol{R}_2\right)$$

$$+ \sum_m \left\langle \frac{q_1 e}{\left|\boldsymbol{R}_1 - \boldsymbol{r}_m\right|} \left[E - H_0\right]^{-1} \frac{q_2 e}{\left|\boldsymbol{R}_2 - \boldsymbol{r}_m\right|} \right\rangle + \sum_m \left\langle \frac{q_2 e}{\left|\boldsymbol{R}_2 - \boldsymbol{r}_m\right|} \left[E - H_0\right]^{-1} \frac{q_1 e}{\left|\boldsymbol{R}_1 - \boldsymbol{r}_m\right|} \right\rangle$$

The dielectric response comes about naturally in infinite-order Brillouin-Wigner theory. We were interested in whether this response resulted in a modification of the Coulomb interaction at short range. At long range (under conditions where many atoms and electrons are between the two deuterons), this kind of model reproduces the dielectric response used by Ichimaru.

After studying the problem for a while, we concluded that the screening effect at short range that follows from this model produces a polarization potential of the form

$$V_{pol} = V_o + \Delta \boldsymbol{R} \mathrm{g} \mathbf{M} \mathrm{g} \Delta \boldsymbol{R}$$

where

$$\boldsymbol{R}_1 = \boldsymbol{R}_{cm} - \frac{1}{2}\Delta \boldsymbol{R} \qquad\qquad \boldsymbol{R}_2 = \boldsymbol{R}_{cm} - \frac{1}{2}\Delta \boldsymbol{R}$$

Based on this, we conclude that the dielectric response at short range should be the vacuum dielectric response. We disagree with the results of Ichimaru in this regard.

Proposal for Electron Bombardment of Metal Deuterides

We proposed the possibility of taking advantage of the thermodynamic stability of vacancies in metal deuterides as a way to create metal deuterides with a high defect concentration. The basic idea was to start with a highly loaded metal deuteride such a PdD or TiD_2, and subject it to electron beam irradiation under conditions where the deuterium could be induced to remain in the metal deuteride (for example, by coating the surface with a blocking material, or by performing the irradiation while keeping the temperature very low). In collaboration with M. Swartz of JET Technology, irradiation experiments were performed on pure Ni, and the defect level was analyzed from resistance measurements. It was found that the electron beam irradiation produced a very high yield of what we assumed were Frenkel defects in nickel (as compared to the literature that we had available for other metals). The defects were observed to heal within about 10 hours. These initial experiments suggest that electron beam irradiation of metal deuterides may be a very attractive way of developing materials with high defect concentrations.

Lattice Resonating Group Method

Most theoretical papers on the dd-fusion reaction have made use of the resonating group method or the R-matrix method. The resonating group method assumes a total wavefunction Ψ_T of the form

$$\Psi_T = \sum_j \Phi_j F_j$$

where the internal nuclear states are included in Φ_j, and where F_j is the channel separation factor. The internal nuclear states are presumed to be fixed, and the channel factors are determined from a solution of the coupled-channel equations

$$E\ F_j = \left\langle \Phi_j \left| \hat{H} \right| \Phi_j \right\rangle F_j + \sum_k \left\langle \Phi_j \left| \left(\hat{H} - E \right) \right| \Phi_k F_k \right\rangle$$

We have proposed a generalization of the resonating group method to include interactions with the lattice as a way of including interactions with phonons at the outset in the formulation. We generalize the resonating group method by using an initial wavefunction of the form

$$\Psi_T = \sum_j \Phi_j \Psi_j$$

where Ψ_j now includes the center of mass coordinates for the reacting nuclei associated with channel *j*, and also the center of mass coordinates for the other nuclei in the lattice. Assuming equivalently that the internal nuclear states Φ_j do not depend on the relative separation, the lattice channel functions can be determined from a solution of the lattice multi-channel equations

$$E\ \Psi_j = \left\langle \Phi_j \left| \hat{H} \right| \Phi_j \right\rangle \Psi_j + \sum_k \left\langle \Phi_j \left| \left(\hat{H} - E \right) \right| \Phi_k \Psi_k \right\rangle$$

These equations can serve as the starting point for modeling anomalies in metal deuterides.

A similar generalization of the R-matrix method to include lattice effects is completely straightforward. The total lattice wavefunction in this case can be developed using lattice channel factors $\Psi_T = \sum_j c_j \Phi_j \Psi_j$, and then an appropriate eigenvalue equation for the amplitudes c_j can be developed.

Separation of local and nonlocal degrees of freedom

Within the framework of the lattice resonating group method, one encounters an extremely interesting situation with respect to modeling when one attempts to include the effects of a highly excited extended phonon mode. When the important physics is local, a description in terms of local lattice variables is convenient. The Coulomb repulsion between nuclei at short range and the strong force that mediates the reaction physics are local effects, best described in terms of local position variables. The oscillation of a lattice in a highly excited phonon mode that involves a very large number of atoms is a nonlocal effect, and is best described in terms of phonon mode amplitudes.

So what happens when we consider a problem that involves both the microscopic Coulomb and nuclear interactions, as well as a highly excited delocalized phonon mode which might prove to be important in bringing the nuclei closer together. To model the local interactions, a local description is convenient. To model the nonlocal interactions, an expansion in phonon mode amplitudes is convenient. When both local and nonlocal effects are important on an equal footing, we face serious issues in arranging for a convenient description of the physics.

This problem can perhaps be best understood by considering the expansion of the nuclear center of mass position operator $\hat{\boldsymbol{R}}_i$ in terms of phonon mode amplitudes $\hat{q}_m$

$$\hat{\boldsymbol{R}}_i = \boldsymbol{R}_i^{(0)} + \sum_m \boldsymbol{u}_m(i)\,\hat{q}_m$$

Here the summation over *m* is over the phonon modes, and $\boldsymbol{u}_m(i)$ is the phonon mode direction vector associated with the center of mass position of the *i*th nucleus. To describe the local physics, we would like to base our analysis on the position operators $\hat{\boldsymbol{R}}_i$. To describe a highly excited phonon mode, we would like to base our analysis on the mode amplitudes $\hat{q}_m$. When both are equally important in a problem, either choice proves to be highly inconvenient for the description of one of the parts of the problem.

For such problems, it was proposed in that a hybrid description might prove to be useful. The essential idea is that if only a single phonon mode is highly excited, then it might make sense to treat that mode explicitly as an extended quantum system. If all of the other phonon modes are assume to have thermal excitation, then their combined effect will add up so as to give a local

response to whatever physics is present. This suggests the hybrid description specific for the case of a single highly excited mode in the presence of a large number of other modes that are thermally excited

$$\hat{\boldsymbol{R}}_i = \bar{\boldsymbol{R}}_i + \boldsymbol{u}(i)\hat{q}$$

The local position operator now is made up of a nonlocal piece that takes into account the excitation of a highly excited phonon mode, and an approximately local piece that is made up of the contributions from all of the other phonon modes.

Interpretation of the Chambers experiment

While the experiment of Fleischmann and Pons and the experiment of Jones provided the motivation for looking into the associated theoretical issues, neither experiment provides much input as to what physical mechanisms are involved. Hence we looked for other claims of anomalies that perhaps might be helpful in this regard.

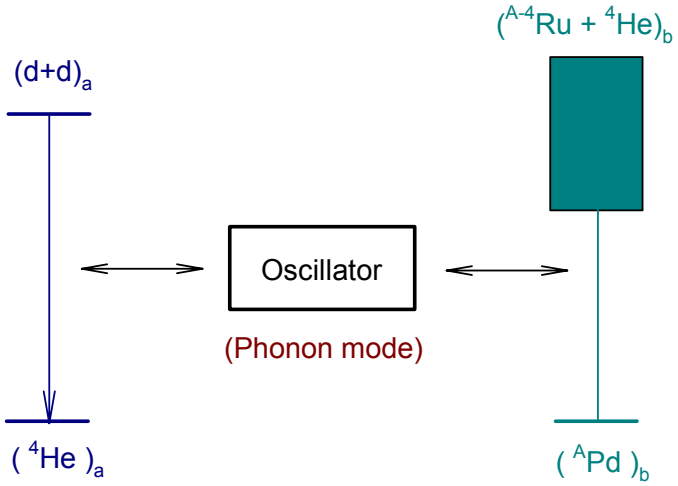

Fig. 2.1: Schematic of second-order two-site reaction for the Chambers experiment. Two deuterons combine to form ^{4}He at site *a*, with the exchange of an even number of phonons greater than 2 to satisfy the microscopic selection rule. The exchange of a single phonon in an electric dipole interaction with an alpha particle within a Pd nucleus at another site results in an ejection at site *b*, with the ejection energy coming from the site *a* reaction.

There are experimental results that appear to require phonon exchange, which we consider in what follows. A very relevant experiment was reported by G. Chambers, then (1990) at NRL, provides one such example. Deuterons in the 500-1000 eV range from an electron cyclotron resonance source were incident on PdD, and alpha emission was detected at low levels between 18-21 MeV. On the one hand, the incident deuteron energy is sufficiently low that it is not obvious that any reactions should have occurred. On the other hand, of all the reactions that might be possible, there are no reactions expected that would yield such energetic alphas.

We have interpreted the result of this experiment in terms of the lattice resonating group formulation as being indicative of phonon exchange. The idea is that phonon exchange by itself is unlikely to have much of an effect on any first-order reaction. However, two reactions that both exchange phonons with a common highly excited phonon mode have the possibility of being coupled in a new kind of second-order reaction. The Chambers experiment is then interpreted as being an example of such a second-order process, where two deuterons fuse to make ^{4}He at one site, while at another site the ejection of an alpha particle from a Pd nucleus is induced as the second part of the process (see Figure 2.1).

The overall second-order two-site reaction is written as

$$\left(d+d\right)_a+\left({}^{A}Pd\right)_b \leftarrow \left({}^{4}He\right)_a+\left({}^{A-4}Ru+\alpha\right)_b+Q$$

The reaction energy Q calculated for this process is 18-21 MeV (there are 6 stable Pd isotopes, each with a different reaction energy as illustrated in Figure 2.2), in agreement with the observation. Very energetic alphas have also been observed in at least 3 other experiments in the past 5 years.

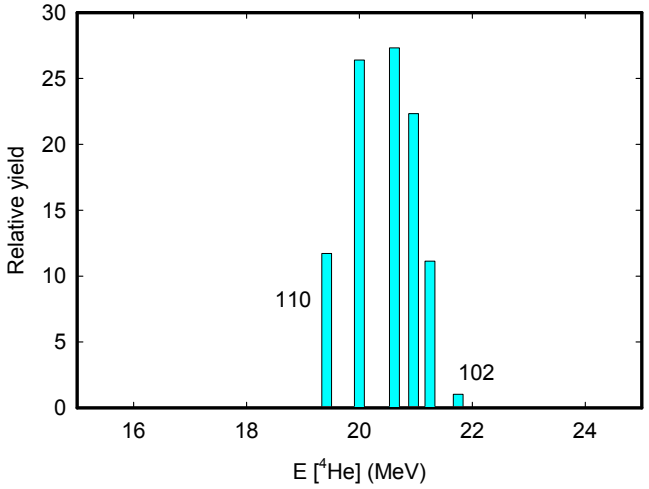

Fig. 2.2: Yield as a function of energy for the different ions assuming transitions to the ground state of the daughter ions.

Second-order Off-resonant Energy transfer

An implication of the proposed explanation for the Chambers experiment is that there exists a second-order off-resonant quantum coupling effect capable of transferring a large energy quantum from one physical system to another through an intermediate physical system characterized by low energy quanta. An interesting feature of this kind of coupling is that the low energy quantum system has the potential of being uniform over locations where two-level systems with a large ΔE occur, which suggests the possibility of a Dicke enhancement of the coupling. This is interesting, and we are not familiar with applications of this effect in the literature. We are concerned both with discrete to continuum transitions [as shown in Figure 2.3] as well as discrete to discrete transitions [as shown in Figure 2.4].

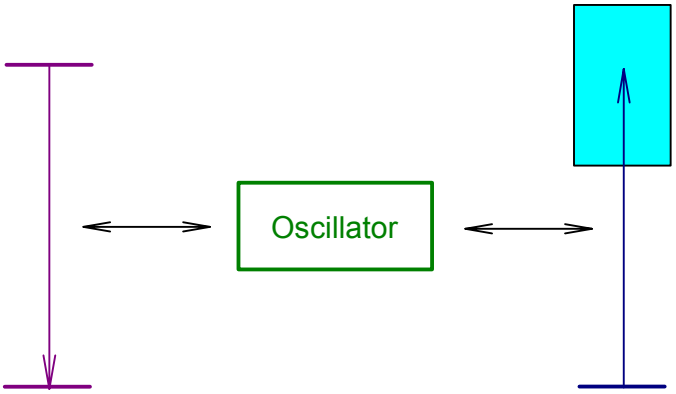

Fig. 2.3: Schematic of off-resonant second-order quantum coupling from a discrete two-level system to a continuum.

The generic experiment that demonstrates an anomaly in metal deuterides presently suffers from disadvantages. Two of these are of interest here: generic poor reproducibility and a lack of easy access to important internal variables. For a theorist, the second problem is by far the more serious. As an example, let us suppose that in the coming months continued work with the Case experiment leads to a moderately reproducible experiment that exhibits both heat and helium which are determined to be consistent with the near 24 MeV fusion reaction energy. We have no hope of making a detailed comparison of theory with experiment at the microscopic level, since we do not know the microscopic lattice structure, overlap probabilities, phononic excitation, or even which phonon modes might be excited, much less to what level.

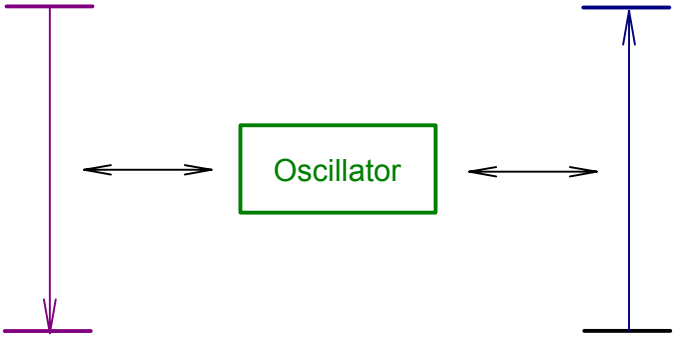

Fig. 2.4: Schematic of off-resonant second-order quantum coupling from a discrete two-level system to a resonant two-level system.

Having proposed an idealized theoretical model that exhibits effects something like what is needed, we are in a position to suggest the possibility of doing experiments with fundamentally simpler systems that are infinitely more accessible. For example, a two-level system is a two-level system, whether it is implemented as a nucleus, atom or spin system. An oscillator is an oscillator, whether it is a phonon mode, a mode of a microwave cavity, or an LC-circuit. Coupling between the different systems can be arranged for by design. We can put atoms in a cavity, and develop quite a good understanding of how strong the interaction is. We can measure the coupling strength directly.

The new models predict new physical effects. Physical implementations of these models can be developed that will exhibit the new physical effects. We can therefore study the essential physics behind the metal deuteride anomalies by using much simpler systems that are optimized for whatever physics that we are interested in. Rather than being at the mercy of a complicated metal deuteride that might or might not work, we can be the master of a simple and well-diagnosed experiment that works every time.

When these ideas were initially proposed, we did not have sufficiently strong tools available to analyze them adequately. Consequently, the initial proposals are perhaps of greatest value as gedanken experiments. More recently, we have revisited the problem of applications that implement some of these ideas. We are presently having some success in applying off-resonant excitation transfer to the problem of thermal to electric energy conversion.

Nonperturbative Analysis of Coupled Systems

When we first proposed applications for the second-order off-resonant excitation transfer effect, we made use of perturbation theory in the analysis of the coupled systems that resulted. It was pointed out by a colleague (R. Rudd) that perturbation theory would be expected to break down under conditions of strong coupling, a result that we confirmed in our modeling. If perturbation theory could not usefully be applied, then some other techniques would be needed. We examined several approaches to this problem.

One approach was to make use of generalized coherent states. In this case, if one has a complicated Hamiltonian $\hat{H}$ that describes a coupled system, and one wishes to evaluate the maximum possible excitation transfer rate, then one proceeds by defining an operator $\hat{Q}$ that measures the degree of excitation transfer. The maximum rate in the case of a time-independent Hamiltonian is

$$\frac{d}{dt}\left\langle \hat{Q} \right\rangle = \frac{1}{i\mathrm{h}}\left\langle \left[\hat{Q}, \hat{H} \right] \right\rangle$$

Consequently, we define an operator $\hat{P}$ according to

$$\hat{P} = \frac{1}{i\mathrm{h}}\left[\hat{Q}, \hat{H} \right]$$

In principle, the maximum rate associated with this degree of freedom could be defined by solving the eigenvalue equations

$$\hat{P}\Psi = \mathrm{h}k\Psi$$

In practice, the eigenvalue solutions are hard to interpret in terms of a system localized in configuration space.

We examined various approaches to generate approximate wave packet solutions in the vicinity of a fixed point in configuration space. One way to do this is to solve instead

$$\hat{P}\Psi + \lambda\hat{Q}\Psi = \mathrm{h}k\Psi$$

where λ can be set to localize the resulting eigenfunction solution where desired. This approach was studied in the thesis of Chris Black. There are related approaches that may also be appropriate. For example, it should be possible to develop eigenfunction solutions for

$$\left[\hat{P}^2 + \lambda\left(\hat{Q} - Q_0 \right)^2 \right]\Psi = \varepsilon\Psi$$

which should also be useful for developing approximate wave packet estimates for reaction rates.

We also examined problems of this sort by direct numerical solution of the associated eigenvalue equation in configuration space. This proved to lead to large numerical errors in the case of simple eigenvalue algorithms and packages that we had available. We looked into implementing stronger algorithms, which allowed us to develop accurate numerical solutions. These algorithms are documented to some degree in the DARPA report.

Finally, we found from the results of the numerical calculations that the different coupled systems that we analyzed had important features in common. All of the models tended to produce eigenfunctions localized to within the relevant coupling strength *g* in the limit that the phonon number and Dicke number became large, almost independent of the details of the coupling associated with the models. This being the case, we proposed to recast the problems in terms of localized basis states corresponding to infinite phonon and Dicke number, but constant coupling strength. Whatever coupling not included in these states was found to produce delocalization, and a theory based on this kind of analysis was built up.

In the end, this approach made clear that delocalized eigenfunctions for the coupled systems in excitation transfer problems only came about as a result of the breaking of the symmetry associated with the coupling coefficients in configuration space. This conclusion was verified numerically as well as analytically. The implications for the metal deuteride models under consideration was that such models would show interesting effects only if they included a mechanism to destroy this symmetry. We note that the site-other-site models presently under discussion do not destroy the symmetry in the coupling coefficients, but do accomplish the same functionality through the inclusion of loss that is highly nonlinear in the configuration space variables.

Site-other-site reactions and the null reaction

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 an 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 must be the “null” reaction, which couples a fusion reaction at one site with the inverse reaction at another site. Such a process is exactly resonant up to the difference in phonon exchange at the two sites. The specific example of this process that we consider to be important for understanding the anomalies in metal deuterides is the two-site null reaction of the form

$$\left(d+d\right)_a+\left({}^4He\right)_b \leftrightarrow \left({}^4He\right)_a+\left(d+d\right)_b$$

A schematic of this reaction is illustrated in Figure 2.5. We presume sufficient phonon exchange (with accompanying angular-momentum exchange) at both sites to satisfy the microscopic E2 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.

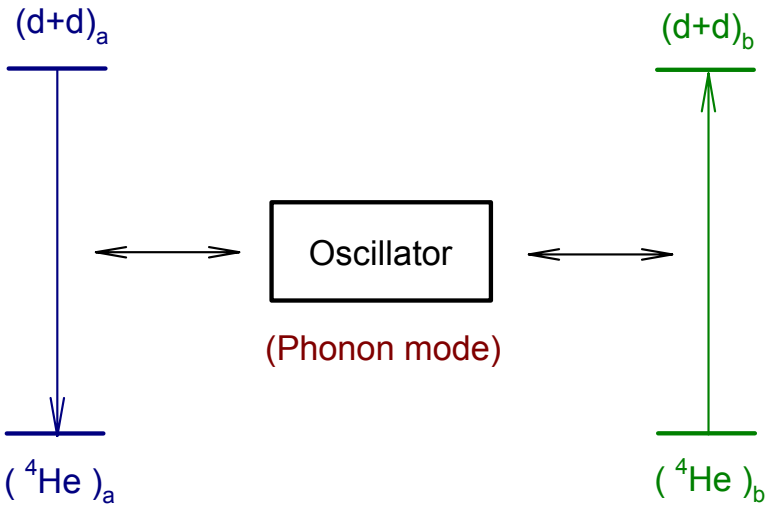

Fig. 2.5: Schematic of the off-resonant second-order sit-other-site null reaction.

Nuclei formed by dissociation have trouble tunneling apart

Of all possible second-order site-other-site reactions, the reaction that should have the fastest associated rate is the "null" reaction

$$\left(d+d\right)_a+\left(^4He\right)_b \leftrightarrow \left(^4He\right)_a+\left(d+d\right)_b$$

In this second-order reaction, essentially nothing happens, other than a pair of deuterons at one site appears to exchange places with a helium nucleus at another site. Yet because the reaction is resonant, it would be expected to be the dominant reaction process of this kind. We wondered for some years whether such a process could be observed. After modeling such reactions, it began to become clear that the two deuterons created from the dissociation of ^{4}He are born within fermis of one another, and that they would have difficulty tunneling apart from one another. Hence it might be possible to observe the reaction perhaps in a collision process in which an incident nucleus interacted with both closely spaced deuterons as part of a single reaction process. The same considerations apply in principle to dissociation to the p+t and n+^{3}He channels.

Interpretation of the Kasagi experiment

Such considerations prompted us to examine an experiment reported by J. Kasagi of Tohoku University in Japan, in which deuterons near 100 keV were incident on TiD. In these experiments, Kasagi noticed that in addition to the conventional dd-fusion products and associated secondary products, there was a very broad fast proton and fast alpha signal. The energetics of the reaction products are not consistent with known reactions, and the very broad energy spectrum appeared to be consistent with a 3-body exit channel. Kasagi proposed that the fast protons and fast alphas were due to a 3-body incident channel given by

$$d+d+d \rightarrow n+p+^4He+Q$$

where the reaction energy Q is about 21.6 MeV. This experiment has been replicated in several labs in Japan and China, and recently in the US at NRL. We have interpreted this experiment as providing experimental support for the existence of the null reaction mechanism. The yield of the reaction in the Kasagi experiment is consistent with 10^{-5} of all of the deuterons being close

together at the fermi scale. More recently, it has become clear that the compact states should include localized t+p and n+^{3}He states as well with energies resonant with the molecular D_2 state.

Compact states

To understand a complicated many-body problem, one usually likes to have a simple analog model, which contains the relevant physics, 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 can be approximated as an exchange potential, leading to

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

where $V(x)$ is the one-dimensional equivalent molecular potential

$$V(x) = \begin{cases} \infty & \text{for} \quad x \le 0 \\ V_0 & \text{for} \quad 0 \le x \le d \\ 0 & \text{for} \quad d \le x \le L \\ \infty & \text{for} \quad x > L \end{cases}$$

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 2.6.

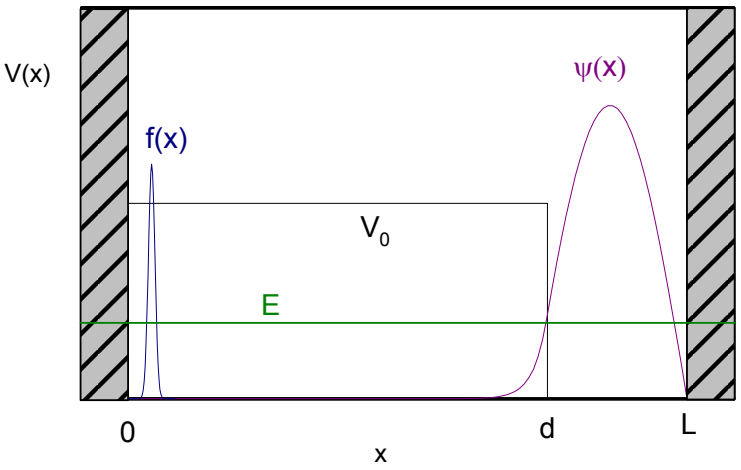

Fig. 2.6: Schematic of 1-D analog model. The molecular potential is modeled by a square well with zero potential between *d* and *L*, and a constant potential below *d*. The unperturbed ground state (analog for the molecular ground state) is illustrated as $\psi(x)$.

This analog model problem is easily solved [see Figure 2.7]. 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 that are produced from helium dissociation, which tunnel apart. We note that this basic argument applies whether the exchange occurs with two deuteron states, or with localized p+t or n+^{3}He states.

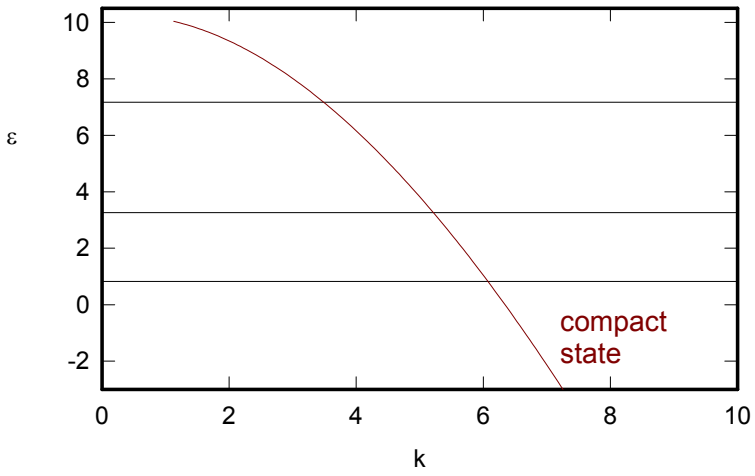

Fig. 2.7: Normalized eigenvalues as a function of the normalized coupling strength *k* for the square well analog. When the coupling strength increases to a sufficiently large value, a new state appears, with an energy that depends of the strength of the coupling. Deuterons that are created at close range try to tunnel apart, but come back together to make helium (sending the excitation elsewhere) before they can tunnel apart.

Compact state angular momentum

Two deuterons in close proximity at the nuclear scale would decay rapidly if there occurs no mechanism to prevent such decay. The analysis of the nuclear interaction in the presence of the lattice indicates that it is possible to exchange many phonons in the event that the relative local motion due to the highly excited phonon mode is greater than 1 fm. Phonon exchange can provide angular momentum on the microscopic scale, and so we consider the possibility that the two-deuteron compact states may have significant angular momentum.

If so, then it is possible that the two-deuteron compact state will be stabilized against decay through the p+t and n+^{3}He fusion channels as the presence of a significant centripetal barrier will inhibit tunneling. We are considering compact state angular momentum on the order of twenty angular quanta or more, which improves the stabilities by tens of orders of magnitude. These considerations apply whether the compact state involves two deuterons, or other localized two-nucleus states such as p+t or n+^{3}He.

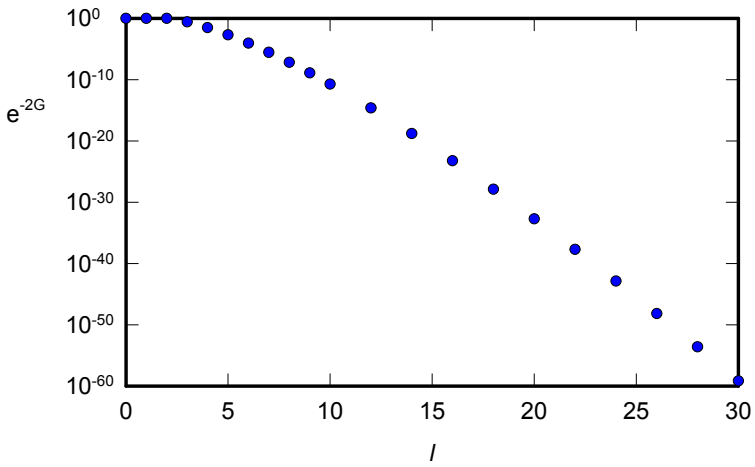

Fig. 2.8: Gamow factor associated with the n+^{3}He channel as a function of angular momentum of the two-deuteron compact state.

This conjecture appears to have experimental support in Kasagi's experiment in two different ways. Firstly, some stabilization is absolutely necessary, as the experimental estimate of 10^{-5} of the deuterium population being in compact states would otherwise result in the loss of all compact state population in a time short compared to a femtosecond. Secondly, the two-body exit channels for the three-deuteron reaction are not observed. The two-body exit channels are

$$d + d + d \rightarrow d + {}^{4}He$$
$$d + d + d \rightarrow t + {}^{3}He$$

A large compact state angular momentum would imply a large exit channel momentum (as a deuteron at 100 keV is not capable of transferring more than one or two units of angular momentum), which would inhibit the two-body exit channels. The three-body exit channel behaves differently, as it is possible to convert between radial and angular momentum in the three-body system.

Proposal for Coincidence Kasagi Experiment

The significance of the Kasagi experiment in light of the discussion here is not appreciated within the scientific community. Most physicists that are familiar with it believe it to be incorrect, in spite of the replications that have been done. It is true that the measurement is not an easy one, due in part to the large background of dd-fusion products.

To address this, we proposed that a coincidence version of the Kasagi experiment be done in which the final state proton and alpha were detected at the same time. An analysis of the problem indicates that an energy-resolved or an angle-resolved version of the coincidence Kasagi experiment should provide very strong evidence in support of the basic result if correct, or else disprove the result convincingly if incorrect. NRL has proceeded with this experiment with support from DARPA, but this experiment has not yet been completed. We are hoping that support can be found to finish this work.

Proposal for Modified Kasagi Experiments

If the interpretation that we have proposed for the Kasagi experiment is correct in that it is detecting a two-deuteron compact state, then variants of the experiment could be developed to measure various properties of the two-deuteron state. An example of such an experiment is

$$\alpha + \left(d + d\right)_{compact} \rightarrow 2\alpha + 23.85MeV$$

As the final state is a two-body channel, this reaction should be suppressed until the incident alpha energy becomes sufficiently large to contribute angular momentum on the general order of the local angular momentum of the compact state. The energy-dependence of the yield could therefore be used in principle to measure this angular momentum.

Many other variants are possible as well. Perhaps most interesting are variants in mixed metal deuterides and hydrides in which the proton-deuteron compact state may be observed.

Model for the Interaction potentials

A typical interaction term that appears in the coupled-channel equations that results from the lattice resonating group method is

$$\left\langle \Phi_{dd}\phi_n Y_{lm} \left| \hat{H} - E \right| \phi_{He}\phi_{n'} \right\rangle$$

We have evaluated this interaction in the case of simple Gaussian (Feenberg) nuclear wavefunctions of the form

$$\Phi_2 = N_2 e^{-\beta_2 |r_1 - r_2|^2}$$

$$\Phi_4 = N_4 e^{-\beta_4 |r_1 - r_2|^2} e^{-\beta_4 |r_1 - r_3|^2} e^{-\beta_4 |r_1 - r_4|^2} \; e^{-\beta_4 |r_2 - r_3|^2} e^{-\beta_4 |r_2 - r_4|^2} e^{-\beta_4 |r_3 - r_4|^2}$$

and a scalar Gaussian Wigner nuclear interaction. The resulting interaction in the WKB approximation is

$$\left\langle \Phi_{dd}\phi_n Y_{lm} \left| \hat{H} - E \right| \phi_{He}\phi_{n'} \right\rangle = -4V_0 \left[\frac{8^{\frac{1}{4}} \left(2\beta_2\right)^{\frac{3}{2}} \left(4\beta_4\right)^{\frac{9}{4}}}{\pi^{\frac{1}{4}} \left(\beta_2 + 2\beta_4\right)^{\frac{3}{2}} \left(\beta_2 + 2\beta_4 + \frac{\alpha}{2}\right)^{\frac{3}{2}}} \right] e^{-K|\bar{r}|^2} \sqrt{2l+1}\,\delta_{m,0}$$

$$\times \frac{1}{\pi} \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} e^{-K|\Delta u|^2 q_{\max}^2 \sin^2 \xi} i_l \left(2K\bar{r} \left| \Delta u \right| q_{\max} \sin \xi\right) \cos\left(\Delta n \; \xi\right) d\xi$$

We are able to evaluate this interaction systematically for different levels of phonon excitation. An example is shown in Figure 2.9. In figure 2.9 we have used

$$v_{lm}^{nn'}(r) = r \left\langle \Phi_{dd}\phi_n Y_{lm} \left| \hat{H} - E \right| \phi_{He}\phi_{n'} \right\rangle$$

We find that a large local amplitude of relative motion can produce are large phonon exchange. We note the linear model outlined here will at some point need to be upgraded, as the relative motion induced by lattice vibration is more likely to be circular.

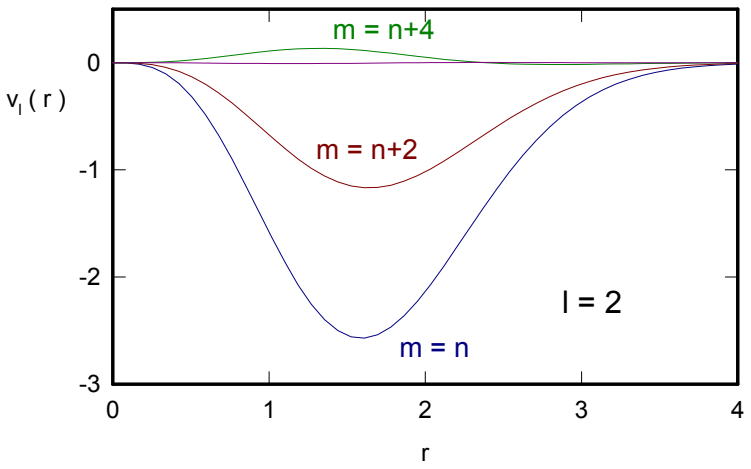

Fig. 2.9: Results for the interaction with model values as given in the text, taking $l = 2$ and assuming that the phonon interaction is characterized by a distance parameter $\Delta u \; q_{\max} = 1\, fm$. The matrix element in this simple model is finite for zero phonon exchange. This is due to a lack of orthogonality in the nuclear states; we expect no $\Delta n = 0$ transitions.

The Two-site Problem

In the analysis of site-other-site null reactions, the most basic problem is the two-site problem. An exchange interaction between reactions at two sites must show up first in the analysis of the two-site problem. We have analyzed in some detail the two-site problem for the null reaction

$$\left(d+d\right)_a+\left({}^4He\right)_b \leftrightarrow \left({}^4He\right)_a+\left(d+d\right)_b$$

in a simplified scalar approximation. We assumed a lattice resonating group state for the nuclear and phonon system of the form

$$\Psi=\sum_n A_n\left|\Phi^a_{He}\Phi^b_{He}\phi_n\right\rangle+\sum_{nlm}\left|\Phi^a_{dd}\Phi^b_{He}\phi_n Y_{lm}\right\rangle\frac{p^a_{nlm}\left(r\right)}{r}+\sum_{nlm}\left|\Phi^a_{He}\Phi^b_{dd}\phi_n Y_{lm}\right\rangle\frac{p^b_{nlm}\left(s\right)}{s}$$

$$+\sum_n\sum_{lm}\sum_{l'm'}\left|\Phi^a_{dd}\Phi^b_{dd}\phi_n Y^a_{lm}Y^b_{l'm'}\right\rangle\frac{p^{ab}_{nlml'm'}\left(r,s\right)}{rs}$$

where the nuclear basis functions $\left|\Phi_{dd}\right\rangle$ and $\left|\Phi_{He}\right\rangle$ are assumed fixed. Implicit in this wavefunction is the assumption of a highly excited phonon mode, accounted for by the presence of harmonic oscillator wavefunctions ϕ_n in the phonon mode amplitude. The remaining radial coordinates are taken to be residual coordinates in the absence of the phonon contribution. Optimization of the radial channel factors in this case leads to the coupled-channel equations

$$EA_n=\left[2E_{He}+\mathrm{h}\omega_0\left(n+\frac{1}{2}\right)\right]A_n+\sum_{n'lm}\int_0^\infty vl^{nn'}_{lm}\left(r\right)\left[P^a_{n'lm}\left(r\right)\right]dr$$

$$E\,P^a_{nlm}\left(r\right)=\left[E_{He}+E_{dd}+\mathrm{h}\omega_0\left(n+\frac{1}{2}\right)-\frac{\mathrm{h}^2}{2\mu}\frac{d^2}{dr^2}+\frac{\mathrm{h}^2l\left(l+1\right)}{2\mu r^2}+V^a\left(r\right)\right]P^a_{nlm}\left(r\right)$$

$$+\sum_{n'}\left[v^{nn'}_{lm}\left(r\right)\right]^*A_{n'}+\sum_{n'}\sum_{l'm'}\int_0^\infty v^{nn'}_{l'm'}\left(s\right)P^{ab}_{n'lml'm'}\left(r,s\right)ds$$

$$E\,P^b_{nlm}\left(s\right)=\left[E_{He}+E_{dd}+\mathrm{h}\omega_0\left(n+\frac{1}{2}\right)-\frac{\mathrm{h}^2}{2\mu}\frac{d^2}{ds^2}+\frac{\mathrm{h}^2l\left(l+1\right)}{2\mu s^2}+V^b\left(s\right)\right]P^b_{nlm}\left(s\right)$$

$$+\sum_{n'}\left[v^{nn'}_{lm}\left(s\right)\right]^*A_{n'}+\sum_{n'}\sum_{l'm'}\int_0^\infty v^{nn'}_{l'm'}\left(r\right)P^{ab}_{n'l'm'lm}\left(r,s\right)dr$$

$$E\,P^{ab}_{nlml'm'}(r,s)=\left[2E_{dd}+\mathrm{h}\omega_0\left(n+\frac{1}{2}\right)-\frac{\mathrm{h}^2}{2\mu}\frac{d^2}{dr^2}+\frac{\mathrm{h}^2 l(l+1)}{2\mu r^2}+V^a(r)-\frac{\mathrm{h}^2}{2\mu}\frac{d^2}{ds^2}\right.$$

$$\left.+\frac{h^2 l'(l'+1)}{2\mu s^2}+V^b(s)\right]P^{ab}_{nlml'm'}(r,s)+\sum_{n'}\left[v^{nn'}_{l'm'}(s)\right]^* P^{ab}_{n'lm}(r)+\sum_{n'}\left[v^{nn'}_{lm}(r)\right]^* P^{b}_{n'l'm'}(s)$$

Of interest here is that we are able to apply the lattice resonating group method now directly to this kind of problem, and develop quantitative results.

The result of the analysis indicates that the localization energy associated with a compact state is several MeV (depending on the details of the interaction), and is greater than the attractive exchange energy for the models that we analyzed. The two-deuteron compact has an energy above the molecular state energy, and is hence unstable against dissociation. Although we had hoped for a stable compact state in the two-site problem, we nevertheless can learn from it. The exchange energy in some of the two-site states is attractive, as would be needed to stabilize the compact state. The only problem is that the attractive exchange potential is too weak to stabilize the compact state in this case. Stabilization occurs in the many-site problem for a sufficiently large number of sites.

In light of the more recent proposal that the compact state can be either p+t or n+^{3}He, this problem needs to be revisited. We conjecture that stabilization of the localized state can occur for compact states made of these pairs, as the lower nuclear energy can be used to offset the localization energy.

The Many-site problem

We have considered the generalization of the problem to the case of many sites. The mechanics of the construction of the many-site coupled channel equations are straightforward, however, the problem seems to be qualitatively richer as we discuss below. The many-site coupled-channel equations are of the basic form

$$E\,P^{\beta}_{M,n,\{l_i\}}(\{r_i\})=\left[E^0_M+\mathrm{h}\omega_0\left(n+\frac{1}{2}\right)\right]P^{\beta}_{M,n,\{l_i\}}(\{r_i\})$$

$$+\sum_i\left[-\frac{\mathrm{h}^2}{2\mu}\frac{d^2}{dr_i^2}+\frac{\mathrm{h}^2 l_i(l_i+1)}{2\mu r_i^2}+V_i(r_i)\right]P^{\beta}_{M,n,\{l_i\}}(\{r_i\})$$

$$+\sum_{\alpha}\sum_{n'}\sum_{j}\sum_{\mathrm{l}j}\eta\left[\beta,\{l_i\};\alpha,l_j,\{l_i\}\right]v^{nn'}_{l_j}(rj)\,P^{\alpha}_{M-1,n',\{l_i\}}(\{r_i\})$$

$$+\sum_{\gamma}\sum_{n'}\sum_{k}\sum_{l_k}\eta\left[\beta,\{l_i\};\gamma,\{l_i\},\overline{l}_k\right]\int_0^{\infty}v^{nn'}_{l_k}(r_k)\,P^{\gamma}_{M+1,n',\{l_i\},l_k}(\{r_i\},r_k)\,dr_k$$

There are a very large number of channels, and it quickly becomes impractical to attempt a direction solution of them. In our previous work, we made use of infinite-order Brillouin-Wigner perturbation theory in order to get some insight as to possible nature of the solutions. Here, we simply note that it appears that such an approach is simply not up to the problem when the

coupling becomes strong enough to be interesting in terms of accounting for the experimental results. Instead, we must make use of alternate approximations.

Of fundamental concern is the question of whether there exist localized solutions to the many-site version of the coupled-channel equations. It seems a priori unlikely that an answer would be forthcoming without a brute force computation on the coupled-channel equations. Our efforts to date on this problem have so far not produced insight. For the purposes of the present discussion, we might adopt as an ansatz the assumption that we can define useful localized states which may or may not be stable, and proceed with the calculation in order to ascertain the goodness of the ansatz with solutions in hand. This appears to be a productive approach.

We propose to simplify matters further in order to allow us to make progress on the development of this very hard problem by assuming that all sites are identical, and furthermore, that the establishment of a localized state at each of these sites will involve the same local superposition of orbitals within the different angular momentum channels. These simplifications lead ultimately to a an approximate time-independent eigenvalue equation based on a Hamiltonian of the form

$$\hat{H} = \Delta E\left(\hat{\Sigma}_z + S\right) + \mathrm{h}\omega_0\left(n + \frac{1}{2}\right) + \left(\hat{\Sigma}_z + S\right)\langle h\rangle + \sum_{n'}\left(\hat{\Sigma}_+ + \hat{\Sigma}_-\right)V_{nn'}\hat{\delta}_{nn'}$$

In this Hamiltonian the $\hat{\Sigma}$ operators are pseudospin operators that are developed as a superposition over Pauli matrices at the different sites

$$\hat{\Sigma} = \sum_i \hat{\sigma}_i$$

The parameter S is the Dicke number for the system. The localization energy for a single site is $\langle h\rangle$, and the $V_{nn'}$ terms are integrals of the interaction potentials and localized orbitals summed over the different angular momentum channels.

We have encountered such a Hamiltonian previously, before we had considered the possibility of localized two-deuteron states, as perhaps applying to a many-body version of the problem in which molecular states would make phonon-mediated transitions to helium states. In that case, the hope was that the number of sites involved would be sufficiently large that the Dicke enhancement could offset the Gamow factors. Here, we apply the Hamiltonian now to the situation where compact states are making transitions, in which case there is no Gamow factor, and the coupling is very strong. In our previous work, we studied this kind of model in order to understand under what conditions such a model might lead to extended states that were sufficiently broad in *n* so as to allow coherence between the states with different number of fusion events and vastly different phonon number such that approximate energy conservation occurred. We were astonished at how this model stubbornly insisted on producing localized states in which the number of phonons exchanged was on the order of the associated dimensionless coupling constant. This being said, we are aware that the eigenfunctions of this Hamiltonian are generally not overly interesting in regards to relating to the physical problem in question, without further input to the problem.

Quantum Flow Models

The basic problem with the model Dicke Hamiltonian lies in its high degree of symmetry when *n* and *S* are large, and *M* is small. In order to develop delocalized solutions, the symmetry needs to be broken somehow. Either we require coupling coefficients that depend strongly on *n* or *M*, or else we need some kind of additional potential that is highly nonlinear in one or both of these quantum numbers. It is the case that working in the limit where the number of helium nuclei that

interact is on the order of the dimensionless coupling strength makes a difference with respect to delocalizing the solutions in *n* and *M* space.

There is another effect which is much more important, and which has a very strong dependence on *M*. This includes loss terms. For example, when two deuterons fuse in the many-site problem, the off-resonant energy ΔE (24 MeV) is more than enough to fuel recoil between localized deuterons and many other highly energetic decay modes. The presence of such decay modes completely destroys the underlying symmetry of the problem, and produces significant delocalization of the wavefunction in *n* and *M* space. Unfortunately, the inclusion of decay channels into a Hamiltonian is not particularly straightforward. Such problems in other disciplines are often handled using density matrices. We wish not to adopt such a formulation here, as the associated complications would likely make further progress more difficult due to the added complexity of the approach.

We are now in a position to isolate the part of the solution that describes the intermediate sector part of the problem. We are interested in solutions to the time independent Schrodinger equation

$$E\Psi = \left(\hat{H}_0 + \hat{V}\right)\Psi$$

We divide up the solution Ψ in terms of sector-dependent pieces

$$\Psi = \Psi_1 + \Psi_2 + \Psi_3$$

where each of the different components is restricted to basis states within the different sectors. The Schrodinger equation can be split up by sector, so that we may write

$$E\Psi_1 = \hat{H}_1\Psi_1 + \hat{V}_{12}\Psi_2$$

$$E\Psi_2 = \hat{H}_2\Psi_2 + \hat{V}_{12}\Psi_1 + \hat{V}_{23}\Psi_3$$

$$E\Psi_3 = \hat{H}_3\Psi_3 + \hat{V}_{32}\Psi_2$$

As in the case of infinite-order Brillouin-Wigner theory, we can eliminate the sink channels algebraically according to

$$\Psi_3 = \left[E - \hat{H}_3\right]^{-1}\hat{V}_{32}\Psi_2$$

The intermediate state equation now becomes

$$E\Psi_2 = \hat{H}_2\Psi_2 + \hat{V}_{23}\left[E - \hat{H}_3\right]^{-1}\hat{V}_{32}\Psi_2 + \hat{V}_{21}\Psi_1$$

Golden rule decay terms arise associated with poles of the resolvent operator, which leads to loss of probability amplitude from the intermediate sector. This probability amplitude is replaced by source terms from the source sector. In the end, we arrive at a flow-type solution

$$\Psi_2 = \left[E - \hat{H}_2 - \hat{V}_{23}\left[E - \hat{H}_3\right]^{-1}\hat{V}_{32}\right]^{-1}\hat{V}_{21}\Psi_1$$

This formulation is what we have in mind to use in place of a density matrix approach.

The formulation of the problem in this way leads to restricted Hamiltonians which are Hermitian, as well as terms that describe transitions between the different sectors, which are not Hermitian with respect to a sector (although Hermitian with respect to the total Hilbert space). To capture this, we propose the definition of an operator $\hat{K}$ defined according to

$$\hat{K}_2 = \hat{H}_2 + \hat{V}_{23}\left[E - \hat{H}_3\right]^{-1}\hat{V}_{32}$$

which is explicitly non-Hermitian with respect to the intermediate sector, in that it includes loss terms. The intermediate solution written in terms of this non-Hermitian operator is

$$\Psi_2 = \left[E - \hat{K}_2\right]^{-1}\left(\hat{V}_{21}\Psi_1\right)$$

In the end, we have arrived at a suitable flow formulation directly as a consequence of splitting up the Hilbert space into source, sink and intermediate sectors.

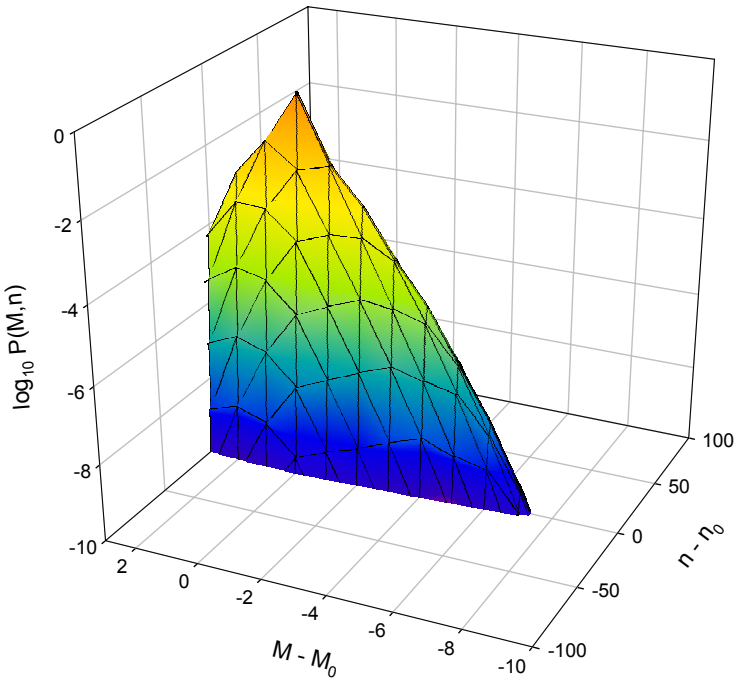

Fig. 2-10: Probability distribution in the vicinity of the source in the case of weak coupling.

Results of Quantum Flow Calculations

We have put together a computer code to analyze the intermediate state solutions along the lines outlined above. Let us consider a few examples in order to illustrate some of the systematics. In

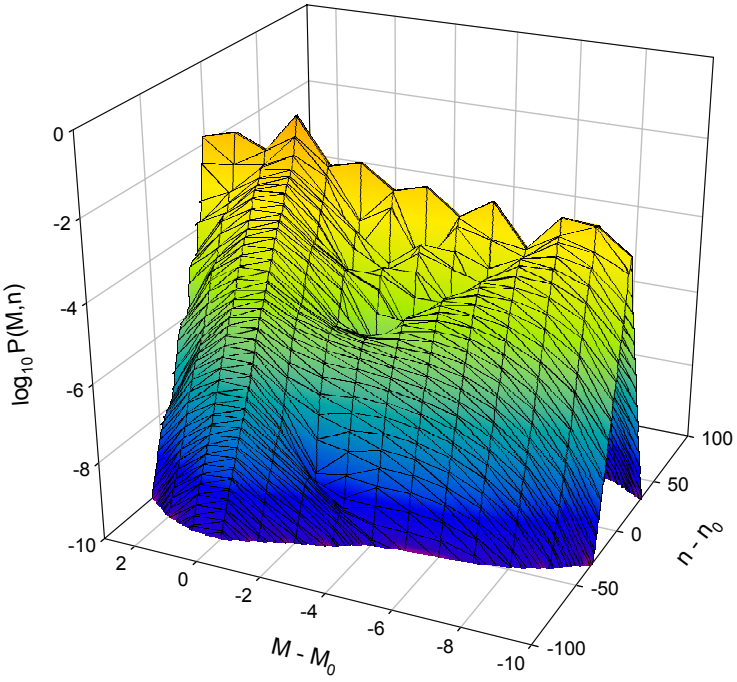

Fig. 2.11: Probability distribution in the vicinity of the source in the case of strong coupling.

Figure 2.10, we show the logarithm of the probability distribution under conditions where the source is localized at (M_0,n_0), and the coupling is weak. In this case, the initial condition corresponds to 3 helium atoms and 10 deuteron pairs. We see that the associated probability density is closely centered around the source, that the distribution is localized in phonon number, and that there is a spread in M that is perhaps larger than one might expect. In the direction of negative M-M_0, which corresponds to more helium nuclei present, the states are very unstable, and the probability distribution decays moderately. The balance between the coupling strength and the decay rate determines the slope. In the other direction, we quickly reach the boundary at which all of the helium nuclei have dissociated, where there is a wall. These states are stable, as they are in serious energy deficit. Such a distribution corresponds to a low or modest level of conventional dd-fusion events, as well as some events in which the fusion energy is transferred to other decay modes within the lattice.

In Figure 2.11, we illustrate the same situation, except that the phonon oscillation amplitude is larger, and the interaction strength for phonon exchange is greater. We see that the stronger coupling leads to a much larger spread in n, which is a hallmark of this kind of model. The spread in M is very significant as well, more so than in the previous example. This spread would like to be even larger, however, in both the positive and negative directions, the distribution hits walls as the number of helium nuclei and deuteron pairs is limited. We see that there is some avoidance of high loss regions of the configuration space, but that this is not a dominant effect in this problem.

In Figure 2.12, we present the logarithm of the probability distribution in the case where there are more helium nuclei present, and the losses are lower (corresponding to the development of higher angular momentum states). We see that the spread in phonon number is now much greater. We see another effect that is of great interest as well. We see that the probability distribution is strongly skewed into the region in which M-M_0 is positive, avoiding the region in which M-M_0 is negative. The avoided region is where deuterons have fused to helium, and where the system has more energy than the local basis state energy, and hence where many decay

processes are allowed. The probability distribution is seen to be favoring low-loss regimes, and hence minimizing the overall loss. This is very interesting, and appears to be a fundamental characteristic of this quantum system.

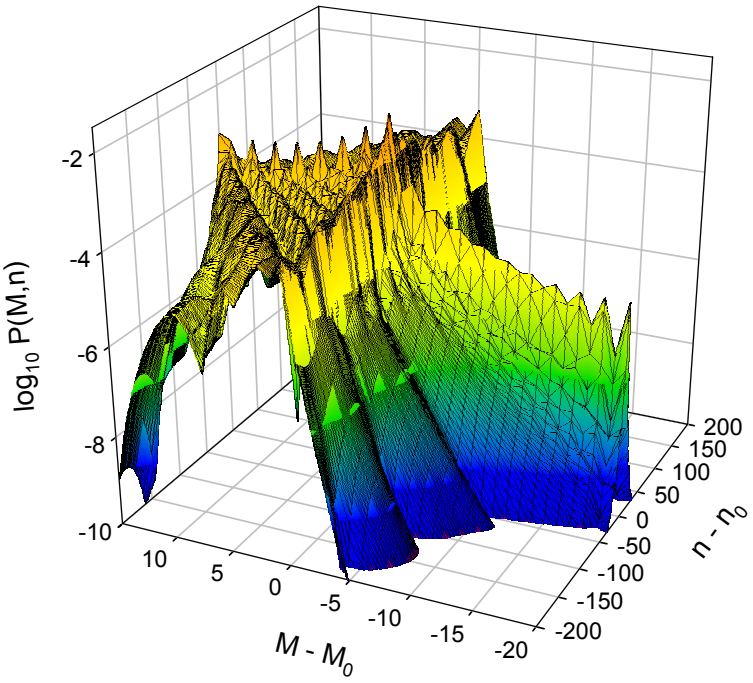

Fig. 2.12: Probability distribution in the vicinity of the source in the case of strong coupling. Only a restricted range in n-n_0 has been included in the plot.

The spread of the distribution in phonon number increases as the strength of the coupling, and decreases under conditions in which the loss is large. It is possible to develop some intuition from these results as to how this problem works. The part of the Hamiltonian that describes fusion and dissociation transitions in this context serves as a kind of kinetic energy operator for the problem. The solutions appear to be outwardly oscillatory away from the source. As long as the probability amplitude avoids lossy regions, then there appears to be a flow from the source into the positive M-M_0 corridor, confined on one side by a wall, and on the other side by an impedance mismatch associated with a high loss region. This flow is increased by a stronger coupling between the states, and inhibited only by the boundary loss. Our calculations so far have indicated that the transport of probability amplitude through the corridor can easily extend for more than a thousand quantum numbers in n-n_0. Altogether this is indicative of a rather efficient mechanism for coupling excitation and energy between the nuclear and phonon degrees of freedom.

Conversion of Nuclear to Lattice Energy

The flow model was developed in order to break the high degree of symmetry in the associated Dicke models with loss terms that are extremely nonlinear in the Dicke number as described above. In the absence of loss, the probability distributions are localized, corresponding to a modest mixing of the nuclear and phononic degrees of freedom. In the presence of loss, the symmetry is broken, and we see massive mixing between the nuclear and phononic degrees of freedom.

With such strong coupling between the different degrees of freedom in the flow model, we began some exploratory computations into the possibility that a large nuclear quantum might be

converted into a large number of small quanta of the highly excited phonon mode. In the physical system, the conversion is

$$\Delta E = \eta \, \mathrm{h}\omega_0$$

where η is on the order of 10^9. We have not yet developed models capable of exploring this region. However, it seemed reasonable to begin considering toy models in which η was assumed to be an adjustable parameter. We have explored quantum flow models with values of η in the range of 100 to 2500, in order to see how the energy coupling varied as a function of η.

Exploratory computations seemed to suggest that the Dicke model augmented with loss leads to very substantial coupling between the nuclear and phonon degrees of freedom that depends very strongly on the size of the "corridor" of stable states away from where the loss occurs. The dependence of the flow solutions on η appears to be very weak, such that the solutions for different values of η over the full range tested were very similar under low loss conditions. An example of such a computation is illustrated in Figure 2.13.

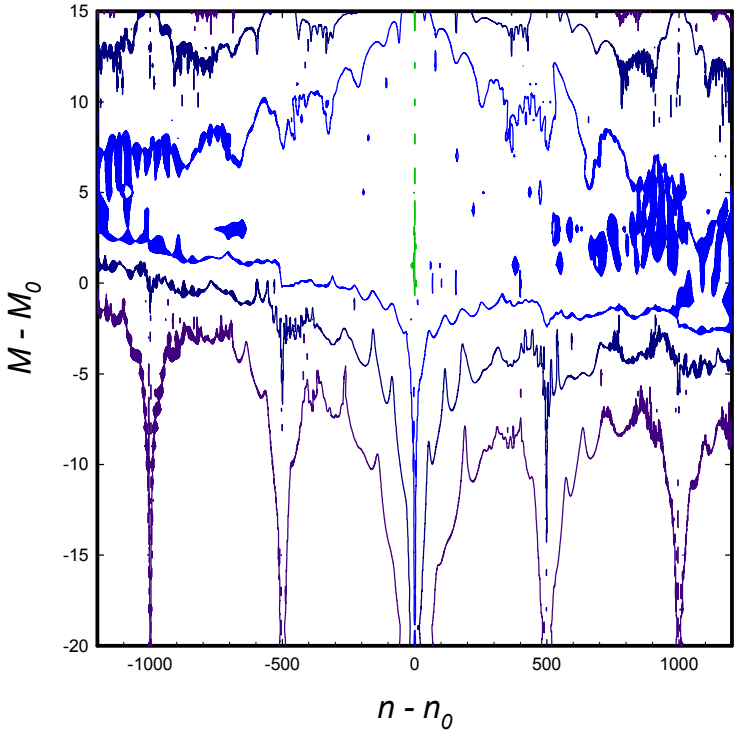

Fig. 2.13: Contours of flow probability for nuclear to phonon energy conversion in a model calculation with η taken to be 500. The probability amplitude is injected at M_0 and n_0, and then couples to nearby values of Dicke number M and phonon number n. The corridor above the equal energy line $M - M_0 = \eta(n - n_0)$ is stable, and allows for the transport of probability to regions of large Δn.

Excess Heat and Size Dependence

In the coupled nuclear and lattice models described in the previous sections, the number of phonons present is assumed to be sufficiently large that additional phonons do not alter the coupling strength. This assumption produces nearly symmetric distributions in n-n_0 on either side of the origin. Hence it is as likely in such models for nuclear energy to be converted to phonons as it is for phonon energy to be converted back to nuclear energy. Such is the coupling generally between different degrees of freedom that become mixed in a coupled quantum system.

We have been interested in the question of whether there might arise some directionality in the coupling, assuming that some way can be found to break the associated symmetry in the problem. The obvious candidate is to arrange for the coupling coefficient to vary as a function of phonon number, as would be the case if the total number of phonons present is on the general order of the number of phonons generated through the conversion of a nuclear quantum

$$\eta = \frac{\Delta E}{\mathrm{h}\omega_0}$$

In this case, the coupling strength increases in the direction of larger phonon number, which draws the injected probability in the same direction. Exploratory computations that we have done so far support this idea.

The corresponding physical situation requires some discussion. Directing the probability flow requires there to be fewer phonons present numerically. However, stable two-deuteron compact states requires there to be as large of an amplitude as possible. Hence the two requirements might seem to work against each other. However, if the total number of atoms participating is restricted to a total number on the general order of η, then the excitation can be strong while at the same time the total number of phonons present is reduced. This argues for the conjecture that the heat effect should proceed at a faster rate in restricted volumes of PdD or of other metal deuterides.

Impact of Resonant Compact States on Tunneling

One of the key questions that remained unanswered in 1989 was the issue of tunneling between deuterons in metal deuterides. For example, the low-level fusion effect of

Jones requires an increase in the tunneling rate over the case of molecular D_2 by tens of orders of magnitude. From theory, such an effect is much too large to be due to modifications of the screening between deuterons in metal deuterides. From experiment, the results of deuteron beam experiments in TiD at low beam energy (a few keV) indicates that the screening in TiD is actually pretty close to the molecular D_2 result. Hence whatever is going on must be due to some other mechanism.

In the course of our work on the two deuteron compact state, we have found that the existence of compact two-deuteron states in the many-site model that are nearly resonant with the molecular state energy can have a profound impact on the radial wavefunction of the molecular state within the metal deuteride.

We have examined this using a radial molecular model augmented with an exchange term at small separation to account for the null reaction exchange process

$$E\,P(r) = \left[-\frac{\mathrm{h}^2}{2\mu}\frac{d^2}{dr^2} + V(r) \right] P(r) - K f(r) \int P(s) f(s)\, ds$$

The strength of the exchange term is determined by the coupling constant *K*, which increases as the compact state becomes more resonant. The impact of the exchange term is illustrated in Figure 2.14. One sees that the probability amplitude at large radius remains unaffected. However, the effect at small *r* is akin to a modification of a boundary condition near the origin. The probability for deuterons initially separated at the Angstrom scale to tunnel to the fermi scale can be increased dramatically.

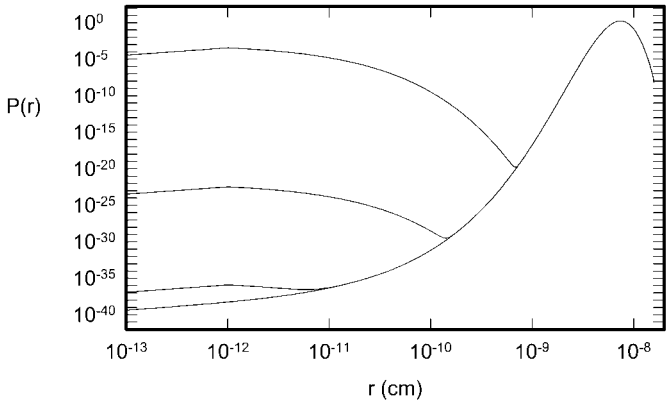

Fig. 2.14: Solutions to the radial molecular D_2 problem with exchange terms of different strength.

Conjecture on the Mechanism for Wolf's Lattice-induced Radioactivity

In 1992, Kevin Wolf at Texas A&M University observed the apparent activation of three Pd cathodes during experiments investigating neutron emission in Fleischmann-Pons cells. That the activation occurred was found about 45 days after the event during an inspection of the data for neutron emission. It was noted that on three occasions, following apparent neutron bursts a large gamma signal was present that the neutron detectors had to reject. The cathodes were placed in front of a gamma detector and found to have significant gamma activity.

That three cathodes are now radioactive is presumed to be fact. The unstable isotopes that were identified are hard to account for. How these isotopes came to be in the cathode is presently not understood. Efforts to replicate the effect in Wolf's laboratory and elsewhere have failed. Measurements of cathodes from many similar experiments around the world have been done, and in no case have others observed such an effect.

We have conjectured that the radioactivity is due to lattice-induced beta decays as a result of second-order two-site reactions of the form

$$\left(p+d\right)_a+\left({}^A Z\right)_b \rightarrow \left({}^3 He\right)_a+\left({}^A\left(Z+1\right)+e\right)_b$$
$$\left(d+d\right)_a+\left({}^A Z\right)_b \rightarrow \left({}^3 He\right)_a+\left({}^A\left(Z+1\right)+e\right)_b$$

and variants of this kind of reaction for electron capture and positron emission. As the p+d reaction is exothermic with more than 5 MeV, we require either fractionation of the nuclear quantum or else partial conversion of the energy to heat in connection with the weak interaction to obtain the roughly 350 keV energy transfer required in the model for the Wolf effect. A similar argument is implied for the case of d+d reactions as well.

This conjecture is speculative, and it can be argued that such an analysis as presented here is premature until the effect can be reproduced. Nevertheless, it is a relatively straightforward exercise to use the data to parameterize a lattice energy transfer model, and then use it to check for consistency. The results of the analysis are interesting (some of which are illustrated in Table 2.1). Many of the observed unstable isotopes can be matched to a single lattice-induced beta decay model, and for the most part isotopes determined experimentally to be absent are also absent in the model. There is general agreement between the model and experiment for roughly 40 such reaction pathways that were examined. Two unstable isotopes are expected from the model in large quantities where none are observed. The model and data in the present form must be understood further before it could be considered to be a completely satisfactory

explanation. Nevertheless, the results are very encouraging, and this approach appears to be promising.

Unstable Product	Stable parent	ΔE (MeV)	n_j (model)	n_j (exp't)	$T_{\frac{1}{2}}$
^{99}Rh	^{99}Ru	2103	0	2.74×10^{9}	15 d
^{100}Pd	^{100}Ru		--	7.7×10^{9}	3.6 d
^{101}Rh	^{101}Ru	541	(both)	1.19×10^{10}	3.3 y
^{101m}Rh	^{101}Rh		4.7×10^{10}	2.21×10^{10}	4.3 d
^{102}Rh	^{102}Pd	1078	(both)	1.21×10^{10}	2.9 y
^{102m}Rh	^{102}Pd		2.9×10^{10}	6.33×10^{9}	206 d
^{103}Ru	^{103}Rh	767	6.0×10^{9}	3.67×10^{9}	39 d
^{105}Ag	^{105}Pd	1348	1.7×10^{10}	2.62×10^{10}	41 d
^{106m}Ag	^{106}Cd	290	5.3×10^{9}	4.51×10^{9}	8.5 d
^{110m}Ag	^{110}Pd	997	$[1.2 \times 10^{13}]$	7.87×10^{8}	252 d

Table 2.I: Model results for the production of unstable isotopes compared with experiment. The lack of agreement for ^{110m}Ag is due to large uncertainties about how this state would be populated from a lattice-induced beta decay from ^{110}Pd. The situation with ^{99}Rh is not understood at this time. The identification of ^{100}Pd is in question due to the absence of ^{100}Rh decay.

Deuterium flux and optical phonons

Almost from the beginning, our interest in theoretical explanations for the anomalies was focused on interactions between nuclei and phonons. Also from the earliest period, it seemed clear from theory that if this were the case that having a more highly excited phonon mode should make the effect stronger. Consequently, we put in some effort trying to understand how to make optical phonons, and also where optical phonons might originate in experiments showing positive results.

We concluded that there were a variety of mechanisms that could be used to generate significant optical phonon excitation. For example, stimulating the surface of a metal deuteride with THz radiation matched to the PdD optical phonon spectrum (in the range of 8-16 THz) is an obvious candidate. To our knowledge, this has not yet been pursued experimentally. We also proposed using the beating between optical lasers mismatched in the THz as a way to couple energy into the optical phonon modes.

However, no such sources were present in experiments early on, and we wondered as to what mechanism might generate optical phonons. Ultimately, we conjectured that optical phonons should be generated when a deuterium flux inside a metal deuteride passes through a discontinuity in chemical potential (illustrated schematically in Figure 2.15). In the PdD experiments, neighboring crystallites have different loadings, which gives rise naturally to mismatches in chemical potential. We proposed fluxing through metal bilayers as a way to increase the chemical potential mismatch. We examined the production of molecular deuterium from the PdD surface under conditions where the loading was sufficiently high so as to produce exothermic desorption.

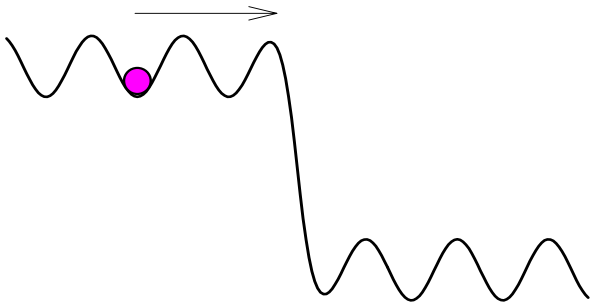

Fig. 2.15: Schematic of deuteron fluxing through a step down in chemical potential.

The earliest evidence in support of this conjecture came from excess heat experiments at SRI in which the excess heat effect occured simultaneously with oscillations in the loading. The excess heat effect was found to increase in the presence of the loading oscillations, and this was interpreted as providing evidence in support of the mechanism under discussion. Subsequently, many experiments have indicated the presence of anomalies correlated with a deuterium flux. The Preparata experiment is based on the development of a large axial current in a long electrode, which drives a deuterium flux through the Coehn effect, and appears to give excess heat and nuclear emissions. Li presented results from an experiment done at Tsinghua University that appears to show an excess heat effect correlated with the fluxing of deuterium through a thin Pd foil. The recent Iwamura transmutation experiment appears to require deuterium fluxing in order to produce the anomaly.

Conjecture Concerning the Iwamura Experiment

Iwamura has recently claimed the observation of a large-scale transmutation effect that has attracted much attention. In this experiment, deuterium is fluxed through a Pd "complex" composed of alternating thin layers of CaO and Pd, with a surface coating of Cs in one case and Sr in another. In situ analysis of the surface Cs over the course of an experiment indicated a possible transmutation of Cs into Pr. Similarly, Sr appeared to transmute into Mo. Isotopic analysis of the product nuclei suggested that the results were consistent with a ^{8}Be increase in the target nuclei.

This result appeared to be relatively solid, in spite of the apparent unlikeliness of the result. In light of the general approach described in this report, we conjecture that what might be going on may be similar to experiments reported previously. We suppose that the fluxing of deuterium through the multilayer structure generates optical phonons efficiently, leading to two-deuteron compact state formation. The compact states are presumed to be reasonably stable. One possible site-other-site reaction process involving the calcium in the thin layers is

$$\left(d+d\right)_a+\left(^{40}Ca\right)_b\rightarrow\left(^{4}He\right)_a+\left(^{28}Si+^{12}C\right)_b+13.35MeV$$

The ^{12}C produced in this way will be energetic (9.35 MeV), such that there is some chance of it reacting with target nuclei in reactions of the form

$$^{A}Z\left(^{12}C,\alpha\right)^{A+8}\left(Z+4\right)$$

To test this hypothesis, one might seek detection of fast ^{12}C nuclei.

We note that ^{12}C might also be created through site-other-site reactions involving oxygen in the CaO layers, predominantly the oxygen reaction

$$\left(d+d\right)_a+\left(^{16}O\right)_b \rightarrow \left(^4He\right)_a+\left(^4He+^{12}C\right)_b+16.69MeV$$

The lattice-induced dissociation in this case should be faster due to the reduced Gamow factor. However, the final ^{12}C energy would only be 4.17 MeV. Such a low energy would lead to a much lower tunneling factor for reactions with mid-*Z* nuclei.

Conjecture Concerning the Letts Experiment

A remarkable result has been claimed by Letts (at the 2003 March APS meeting) in a electrochemical experiment involving a specially prepared Pd cathode with gold on the surface appears to show a dramatic gain in the excess power when stimulated by an optical laser at low intensity. In this case, red lasers in the range of 1-30 mW have given excess power gains up to two orders of magnitude greater than the input power of the laser.

These experiments were initiated in an effort to stimulate the optical phonons through a beating effect as mentioned above. When positive results were obtained, we proposed as a test to clarify the physical mechanism that the test be repeated with only a single laser in order to determine whether the beating effect was responsible, or whether the optical laser was causing the effect. It was found that the effect was present when only a single laser was used, which showed that the beating effect was not necessary to produce the increased excess heat effect. This experiment has been successfully replicated in at least four different laboratories.

The implication of the experiment in light of the theoretical models under discussion requires some thought, as the frequency of the laser is not matched to the optical phonon spectrum. We have conjectured that in metal deuterides there is a very strong mixing between the optical phonon spectrum and the electron plasmon modes, such that laser stimulation results in locally coherent surface mixed-mode excitation at optical frequencies that results in a contribution to the relative motion between deuterons in close proximity.

Improved Nuclear Models

DARPA funds were used to support a PhD student looking into the basic problem of phonon and nuclear coupling based on more sophisticated models. It was felt that following substantial exploratory work on the problem that it was time to try to improve the models by taking advantage of more sophisticated nuclear wavefunctions and interaction potentials. We were interested in providing theoretical support for the coincidence Kasagi experiment at NRL. In addition, Letts is claiming that there is a significant impact of the laser-induced excess heat effect in the presence of a magnetic field. This result would perhaps indicate that nuclear spin effects are important in the problem if the claims are correct.

To proceed, we require correctly antisymmetrized RST few-nucleon wavefunctions that are eigenfunctions of the symmetric group. We have devoted a significant effort toward the application of group theory to the systematic development of such wavefunctions, which has recently led to a complete set of wavefunctions for the three-nucleon and four-nucleon problems. These wavefunctions involve a construction of the form

$$\Psi=\sum_j c_j\left[S\right]_j\left[T\right]_j\left[R\right]_j$$

where the $\left[S\right]_j$ are pure spin functions, the $\left[T\right]_j$ are pure isospin functions, and the $\left[R\right]_j$ are spatial functions. These wavefunctions will be used to implement more sophisticated nuclear potential models. We will begin with an analysis of the Hamada-Johnston potential as an example.

References

P. L. Hagelstein, ``Anomalies in Metal Deuterides,'' *Proceedings of the 9th International Conference on Cold Fusion*, Beijing, May 2002.

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3 Nearly-isentropic Energy Conversion with Quantum Excitation Transfer

Sponsors
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Last year we reported on some new ideas about making use of the second-order quantum excitation transfer effect in a thermal to electric conversion scheme. The basic idea in such a scheme is to transfer excitation from a two-level quantum system in a hot element across a vacuum gap to a cold converter, and then make use of a conventional conversion scheme in order to turn the transferred excitation into electrical work. The implementation that we described previously involved the transfer of excitation to resonant interband transitions in a semiconductor. The electron and hole pairs created as a result of the excitation transfer can be converted using a photodiode, as the excitation transfer effect leads to the same result as conventional photoexcitation, but through a different mechanism.

Calculations on this scheme indicate that the basic approach is sound, and that the power per unit area that can be transferred is respectable – although not as high as our initial calculations of last year seemed to indicate. In analyzing the approach, it became clear that the weak link in the scheme was in the relatively weak coupling associated with an interband transition, as the associated dipole matrix element is on the general order of an Angstrom, which reflects the fact that electron and hole generation ultimately involves the promotion of an electron on a single atom. This is in contrast to the case of an intersubband transition, where an electron in an extended quantum well state makes a transition to a different extend quantum well state. The associated matrix element in this case can be two orders of magnitude larger.

This issue was understood initially, however, we did not first study excitation transfer in intersubband transitions because it was not obvious *a priori* what kind of conversion scheme might be appropriate. More recently, we have understood that there do exist schemes that can utilize the energy of an excited electron to do electrical work – that the creation of an electron and hole pair is not required. This realization has opened up the possibility of a new class of devices based on second-order quantum excitation transfer using intersubband transitions, with subsequent single carrier energy conversion. Here we propose to introduce the basic concept.

Excitation Transfer and Entropy in an Idealized Model

We first consider the operation of a highly idealized device that has the great advantage of simplicity, such that we can introduce the underlying ideas cleanly. This example has the unfortunate property that it does not actually convert excitation into electrical work, but this will be addressed shortly. We consider the scheme indicated in Figure 3.1.

In this scheme, one unit of excitation on the hot side on the left is transferred to an electron initially in the ground state of a quantum well on the left hand side of the device. Once the electron is promoted to the excited state (through second-order quantum excitation transfer), it is able to tunnel to the excited state of the quantum well on the right hand side of the device. Once there, we presume that it transfers its excitation to a two-level quantum system on the cold side to the right of the device. We are interested in understanding the different ways such a simple device might operate.

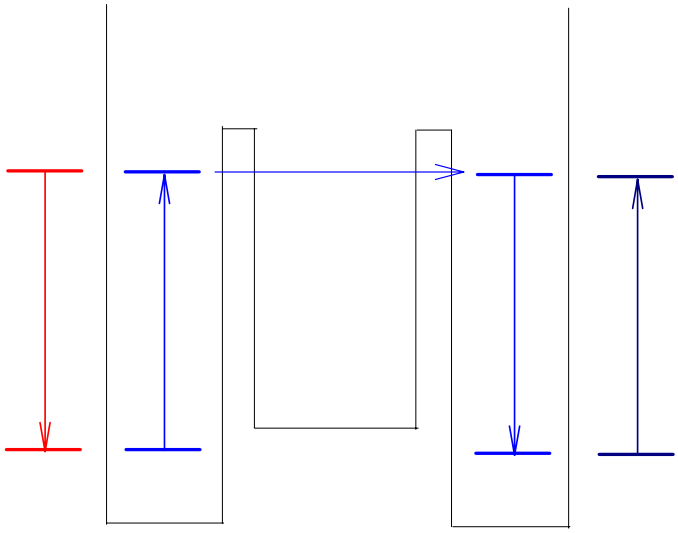

Fig. 3.1: Schematic of simple idealized excitation transfer and tunneling scheme. An initial excitation on the hot side (red) is transferred to an electron in the ground state of the well on the left. The electron is able to tunnel from the excited state to the well on the right. The excitation is then transferred to a two-level quantum system on the cold side (dark blue) to the right of the device.

It is of course possible for the excitation to be transferred back to the hot side immediately after the first excitation transfer event. In this case, the hot side retains whatever energy and excitation, and overall nothing happens. It is possible for the electron to tunnel to the well on the right, and then tunnel back, and then transfer its excitation to the hot side. Once again, overall nothing has changed from the initial condition.

If we next presume that we have many such devices, initialized such that the electron is initially on one side or the other with equal probability, then we are interested in the rate at which forward (hot to cold) transitions occur relative to reverse (cold to hot) transitions. The forward flow is proportional to the number of excited state two-level systems on the hot side. The reverse flow is proportional to the number of excited state two-level systems on the cold side (we will ignore for the purposes of this discussion the possibility of interference of relative level excitation for the second excitation transfer). In such a case, the initial rate of excitation transfer from the hot side to the cold side is proportional to

$$\Gamma \; : \quad \left[\frac{1}{e^{\Delta E/kT}-1}\right]_{hot} - \left[\frac{1}{e^{\Delta E/kT}-1}\right]_{cold}$$

This is interesting in that the forward and reverse flow in this simple example is determined by the temperature on either side, and not by other factors. This is one of the features of a symmetric idealized device such as this one – it becomes easy to visualize the relation between forward and reverse flow with thermal sources and sinks at different temperatures. It is the case that for more complicated structures, the flow associated with individual channels would also be related in this way (because of the principle of detailed balance), but it can quickly become much less intuitive how this works for a highly nonsymmetric device.

Let us consider entropy within the context of a single device. A feature of such an idealized system is that due to the restricted number of states, the overall process conserves entropy. Quantum excitation transfer from one two-level system to another generates no entropy. Tunneling in the absence of scattering generates not entropy. And the second quantum excitation transfer process also conserves entropy. Hence this simple excitation transfer scheme is isentropic. In the event that this general approach can be modified to the problem of energy conversion, perhaps it is possible to move toward a nearly isentropic energy conversion scheme.

Such a scheme would operate near the Carnot limit, since the Carnot limit in this application is obtained directly from entropy conservation.

Energy Conversion

We next consider the application of these ideas to the problem of energy conversion, once again from a highly idealized point of view. We see from the simple device discussed above that an electron flow can be induced through electron promotion on the ``hot side'' (keeping in mind that the device itself may be cold – the coupling with the two-level system on the hot side leads to more electron promotion in the forward direction) followed by tunneling to the other side. The return flow is less if the quantum well levels are coupled to a colder two-level system on the other side.

To extract electrical work, we require that a potential drop be present between the two quantum wells, and we will require in addition the ability to circulate the electrons that flow between the two quantum wells to an external load. The problem is no longer symmetric due to the presence of a potential drop. The requirement of arranging for current flow to an external load also adds complications. Nevertheless, it is possible to devise a relatively simple idealized schematic in this case, as is illustrated in Fig. 3-2.

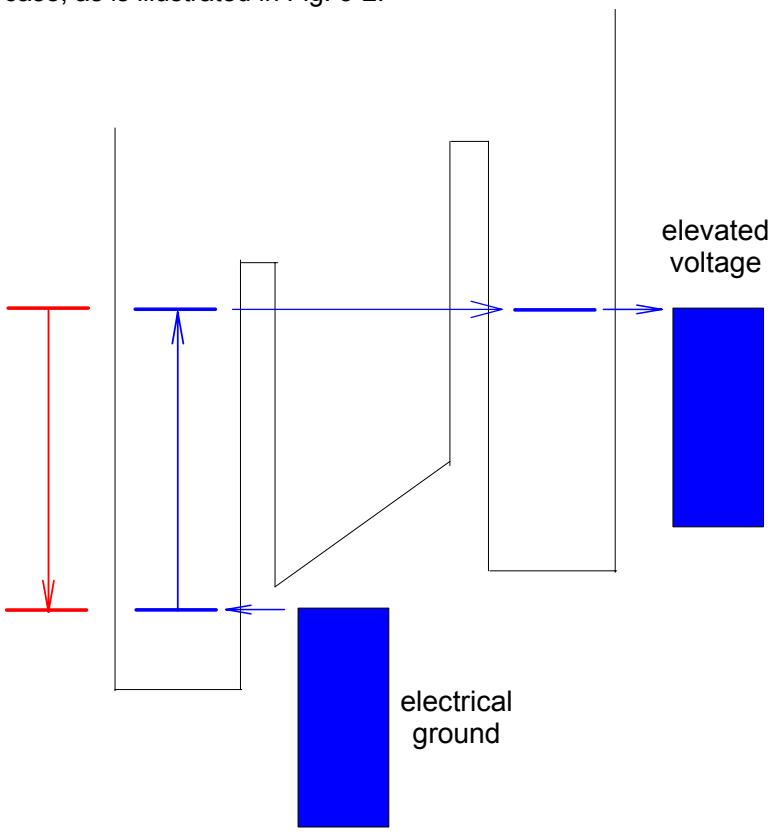

Fig. 3-2: Schematic of a simple energy conversion scheme. Second-order quantum excitation from a two-level system on the ``hot'' side promotes electron excitation in the quantum well on the left. The excited electrons tunnel to a single level in the quantum well on the right. The electron then tunnels to an electrical contact that is connected to a load. The return current path from the load provides electrons for the ground state of the two-level system in the quantum well on the left.

The schematic of Figure 3-2 illustrates the basic idea. Electrons flow into the ground state of the quantum well on the left in close proximity to a ``hot'' two-level system, where second-order quantum excitation transfer promotes them. They tunnel to a single level in a quantum well on the right side of the device. The energy of this level is determined by an external bias, which we have assumed has tuned it onto resonance in the illustration. This single quantum level is assumed to be connected (by tunneling) to an electrical contact at elevated voltage. The electron physically goes from the single level to the contact, and leaves the device. Current flows from the biased contact, through an electrical load, back to the contact noted as electrical ground in the schematic. This contact provides a source of electrons for the ground state of the quantum well.

Once again, the excitation transfer process is isentropic. Electron tunneling (in the absence of scattering) from the well on the right to the well on the left is also isentropic. Hence the electron dynamics within the device in the absence of scattering is isentropic, and achieves potentially Carnot-limited internal conversion efficiency. The connection of the quantum levels with the contacts is assumed to be a source of entropy, one that is not well understood by us at the present.

In this scheme the hot side temperature is fixed by the temperatures associated with the two-level quantum systems to the left of the device. Yet there is no symmetrical equivalent two-level system on the cold side obvious in the schematic of Figure 3-2. Nevertheless, we would expect a return current flow that originates in the contact on the right hand side. As this contact is a thermal reservoir, the associated reverse current flow in this device should be characterized by the temperature of this contact.

Discussion

These ideas represent a major departure from the approaches to the energy conversion problem currently under investigation within the relevant part of the scientific community. There has been no application of the second-order quantum excitation transfer mechanism to this problem outside of our earlier proposals. The notion of a single carrier conversion scheme along the lines discussed here appears also to be new. The thought that it might be possible to achieve nearly isentropic thermal to electric conversion is not under discussion within the community at this time.

We are interested in moving forward to develop experiments that test various aspects of the scheme under discussion. It remains to be demonstrated that excitation transfer can be accomplished with devices of the type implied by the discussion here. Energy conversion with electrons in the absence of holes in such a converter remains to be shown. Hopefully we can begin developing relevant experiments to explore the new ideas in the coming years.

One implication of the new results concerning the thermal diode that is discussed in the first section is the possibility that very efficient solid-state conversion devices should be possible by pursuing such an approach. We would expect that such devices would be limited in the temperatures that they might operate because of thermally induced atomic self-diffusion within the thermal diode. The scheme under discussion here involves a solid-state converter that is to be kept relatively cold (room temperature or perhaps less to minimize scattering effects), and hence is free of this kind of problem. Although we did not pursue the issue here, we would expect the converter to be able to accept energy transfer from hot sources that do not possess well-defined discrete resonant levels.