Phonon models for anomalies in condensed matter nuclear science

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Although excess heat has been studied in the Fleischmann–Pons experiment for more than a quarter century, there is as yet no agreed upon theoretical explanation. Here we divide the problem between known physics issues and new physics issues. In our approach there is an important new physics model which addresses the fractionation of a large quantum; and a new fundamental Hamiltonian which describes the coupling between vibrations and internal nuclear degrees of freedom.

Keywords: Condensed matter, known and new physics, large quantum, phonon models.

Introduction

IN conventional nuclear reactions, one is able to observe energetic reaction products which can be used to develop and understand the microscopic physics at the nuclear scale. In the Fleischmann–Pons experiment[1,2], there are no energetic nuclear emissions in amounts commensurate with the energy produced[3,4]; yet because of the large amount of energy produced, absence of commensurate chemical products, and presence of commensurate amounts of new ^{4}He in PdD experiments[5,6], there seems to be no other conclusion than that the source of the energy must be nuclear. However, whatever nuclear process is responsible for the energy must be fundamentally different than what occurs in conventional nuclear reactions. The absence of commensurate energetic reaction products means that we cannot use conventional tools to study and understand the primary reaction mechanism. Because of this our understanding of the microscopic physics is after a quarter of a century very much incomplete; there is no consensus among those studying the effect as to what reaction mechanism is involved, or even what basic physics is involved. In the brief review that follows, we will outline our approach to the problem, and provide an overview to the progress made so far.

Known physics and new physics

Much of what goes on in the Fleischmann–Pons experiment involves known physics; yet from the comments above clearly new physics is also involved. If we begin with the ansatz that we need to find a way to arrange for deuterons to be in close proximity, then we can understand some aspects of the experiment and observations. The background electron density in PdD due to Pd is too high at the O-sites for D_2 formation, which means that most of the loaded cathode can be considered inert[3,7,8]. Regions of lower electron density are available at defects, with monovacancy being the simplest example. However, the defect density even for the most defective Pd cathode is too low to be of interest. The thought is that Pd vacancies are stabilized at high loading, and become thermodynamically favoured at a D/Pd loading of 0.95 or higher near room temperature[9]. Vacancies do not diffuse near room temperature; so we need to form new metal through codeposition (or inadvertent codeposition at high loading). The active sites in this picture are superabundant Pd vacancies[10] formed on the outer 100 nm skin of cathodes where codeposition has occurred under conditions of high surface loading. This picture is consistent with the (two) loading requirements observed in experiments (maximum D/Pd ratio of ≥0.95 achieved[11] and D/Pd loading of ≥0.85 at the time of excess heat[12]).

If D_2 can interact in a new way to form ^{4}He, then at high power level we expect the active sites to accumulate helium, which inhibits further reactions at the blocked sites. Getting the helium out of the traps associated with the active sites, and diffusing it to a nearby surface is required then at high power level. This picture is consistent with the observed temperature dependence of the excess heat[3,13].

If you have deuterium at the proper loading in superabundant vacancies, excess heat still needs to be triggered. In the models we have studied, the nuclear energy quantum is fractionated into much smaller quanta, which can go into vibrational modes. For this to work in the model, the vibrational modes first need to be highly excited (see refs 14–16). In the Fleischmann–Pons experiment the electrochemical current is associated with the deuterium flux at the surface, and a flux of deuterium through PdD is efficient at generating optical phonon excitation in PdD; hence we expect from this picture deuterium flux to be a trigger[3], as is observed in experiments[17]. Current thinking is that deuterons are responsible in fractionating the nuclear quanta in operation with excited optical phonon modes, and the deuterons can accomplish this cleanly. However, THz acoustic mode excitation would

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also be expected to produce fractionation with participation of the host Pd nuclei, which do not fractionate cleanly (leading to disintegration of the Pd nuclei)[3,18]. Hot spots in PdD are occasionally observed, with associated elemental anomalies; this we attribute to local acoustic mode operation.

New physics and fractionation of a large quantum

We understood in 1989 that excess heat in the Fleischmann–Pons experiment as a nuclear effect without commensurate energetic particles present implied that the large nuclear quantum could be fractionated. The only question is how can this be done? A large number of schemes, models and variants were studied without success until the first sign of success in 2002 (ref. 19), when it was found that the spin-boson model augmented with off-resonant loss could fractionate a large quantum[20].

A weak version of the effect is known in the spin-boson model[21–23]

$$\hat{H} = \Delta E \frac{\hat{S}_z}{\hbar} + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V \frac{2\hat{S}_x}{\hbar}(\hat{a}^\dagger + \hat{a}), \tag{1}$$

in which slow coherent transitions where the large two-level system quantum ΔE is divided into an odd number of oscillator quanta $\Delta n\hbar\omega_0$. We were interested in what limited coherent energy exchange in the multi-photon regime of the model. In perturbation theory the problem can be seen to involve indirect coupling between two distant degenerate states, in which a large number of pathways contribute. The contribution from each pathway individually can be sizeable, even when Δn is large; however, there is strong cancellation between the different pathways[24]. We conclude that fractionation in the spin-boson model is inhibited by a destructive interference effect. Hence a model in which the destructive interference is disturbed can be expected to fractionate a quantum.

One way to accomplish this is to add loss to the system. This can be done by adding a bath which couples to the oscillator according to

$$\hat{H} = \Delta E \frac{\hat{S}_z}{\hbar} + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V \frac{2\hat{S}_x}{\hbar}(\hat{a}^\dagger + \hat{a}) + \sum_j \varepsilon_j \hat{b}_j^\dagger \hat{b}_j + \sum_j U_j (\hat{b}_j + \hat{b}_j^\dagger)(\hat{a}^\dagger + \hat{a}). \tag{2}$$

This model is sufficiently complicated that it is problematic to develop estimates for the coherent energy exchange rate under conditions relevant to experiment directly. We have recently done modelling directly with this model for some 'easy' problems, and we see large enhancements of the indirect coupling matrix element. A simplification of the model can be accomplished using infinite-order Brillouin–Wigner theory to replace the bath with its effect on the spin-boson model to lowest order, leading to a model which can be denoted as

$$\hat{H} = \Delta E \frac{\hat{S}_z}{\hbar} + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V \frac{2\hat{S}_x}{\hbar}(\hat{a}^\dagger + \hat{a}) - \frac{i\hbar\gamma(E)}{\hbar}\hat{a}^\dagger \hat{a} + \hat{a}^\dagger(\hat{a} + \hat{a}^\dagger)F(E) + F(E)(\hat{a} + \hat{a}^\dagger)\hat{a}. \tag{3}$$

The lowest order impact of the bath is to produce loss and fluctuations. In 'easy' problems, this model can give results that agree reasonably well with the full model. We have pursued this approach to the next (third) order in the hope of developing models easier to analyse than the full model. However, we find this approach to have issues in the strong coupling regime, which is required for fractionating a large quantum.

In our early exploratory calculations, we found that a modest disruption of the destructive interference led to an indirect coupling coefficient which was of the same order as if we simply eliminated the lower half of the intermediate states[19]. This led us to focus on a model in which the loss was assumed to be infinitely fast for all basis states with an energy less than the energy eigenvalue (or less than a cut-off energy)

$$\hat{H} = \Delta E \frac{\hat{S}_z}{\hbar} + \hbar\omega_0 \hat{a}^\dagger \hat{a} + V \frac{2\hat{S}_x}{\hbar}(\hat{a}^\dagger + \hat{a}) - \frac{i\hbar\Gamma(\hat{E})}{\hbar}. \tag{4}$$

This model was analysed systematically in the weak, intermediate and strong coupling regimes[24–29]. Scaling laws were established that apply in the large Δn regime, so we can quantify the rate at which a large quantum can be fractionated when Δn is of the order of 10^9 and also understand the requirements to accomplish such fractionation.

More recently, we have developed new approximate models that extend this analysis to more realistic loss-models[26]. Our goal in this was to develop a threshold condition for the onset of fractionation. The intuition was that we should be able to fractionate when the loss is stronger than the spin-boson coupling part of the problem. A corresponding mathematical constraint has been developed; however, in tests against model calculations we have found that it is too conservative. In the most recent calculations we are observing fractionation even with loss models that we consider to be quite weak.

Coupling between nuclei and vibrations

With a possible solution available to the problem of fractionating a nuclear quantum, the question naturally arises as to what coupling mechanism might be relevant. In our interpretation of the Fleischmann–Pons experiment, and of quite a few other experiments, the basic interaction involves vibrations and nuclei. The corresponding oscillator from the spin-boson model might be a highly excited optical phonon mode. The two-level systems would correspond to nuclear transitions; the bath might correspond to conduction electrons, to bound electronic transitions in the host Pd atoms and to nuclear transitions in the host nuclei. An issue then is the coupling mechanism, since according to textbooks the coupling between vibrations and the internal nuclear degrees of freedom is of high order and very weak. We were not able to get numerical agreement between the new models and experiment with existing models for vibrational coupling[20].

We put in a large amount of effort on the problem of coupling between vibrations and nuclei, ultimately with the result that there did not seem to be any coupling sufficiently strong to do the job with conventional mechanisms. We have understood for years that there is a strong coupling between the vibrational degree of freedom and internal nuclear degrees of freedom implicit in a relativistic model, but this coupling is normally eliminated by a generalized Foldy–Wouthuysen transformation. Consequently, we rejected such an approach many times. It was only when we understood that the same issue was present in the spin-boson model, where the first-order coupling can be similarly eliminated with a Foldy–Wouthuysen transformation, and where it is unhelpful when off-resonant loss is present, that we returned to the relativistic model. The fundamental relativistic Hamiltonian under discussion is[30]

$$\hat{H} = \sum_j (\mathbf{M}c^2 + \mathbf{a}\cdot c\hat{\mathbf{P}})_j + \sum_k \frac{|\hat{\mathbf{p}}_k|^2}{2m} + \sum_{j<j'} \frac{Z_j Z_{j'} e^2}{4\pi\varepsilon_0 |\mathbf{R}_{j'} - \mathbf{R}_j|} + \sum_{k<k'} \frac{e^2}{4\pi\varepsilon_0 |\mathbf{r}_{k'} - \mathbf{r}_k|} - \sum_{j,k} \frac{Z_j e^2}{4\pi\varepsilon_0 |\mathbf{R}_j - \mathbf{r}_k|}. \quad (5)$$

If we use a Born–Oppenheimer approximation, then the lattice nuclear problem that remains is[30,31]

$$\hat{H} = \sum_j (\mathbf{M}c^2 + \mathbf{a}\cdot c\hat{\mathbf{P}})_j + \sum_{j<k} V(|\mathbf{R}_j - \mathbf{R}_k|) - \frac{i\hbar\hat{\Gamma}(E)}{\hbar}, \quad (6)$$

where we have augmented the normal Born–Oppenheimer model with a loss term due to coupling with the electrons. This can be reduced to the lossy spin-boson model if we focus on a single nuclear transition. In light of the discussion above, we have in this a starting place to analyse coherent energy exchange between nuclei and vibrations under conditions of fractionation[18,32].

Some effort has been put in to evaluate the phonon–nuclear coupling matrix elements. Coupling in the case of the deuteron has been discussed earlier[33]; of interest in this calculation is that selection rules resulted that may be relevant to enhancements seen in experiments with magnetic fields. We expect that a stronger matrix element will result in the case of coupling to negative energy states, which would have similar selection rules. Coupling in the case of the $D_2/^4He$ transition has also been analysed[34]. The matrix element that results is consistent in magnitude with what is needed to account for the rate at which excess heat is observed in experiments (within the framework of the donor and receiver model of Hagelstein and Chaudhary[35]). It seems plausible that longitudinal mode excitation is favoured in the two-laser experiments because the tunnelling matrix element would be enhanced with local compression of dideuterium.

Discussion

We can make use of the ideas above to consider now the new physics involved in excess heat production in the Fleischmann–Pons experiment[3]. Fractionation is easier when fewer oscillator quanta are involved; so we would expect the highest frequency vibrational modes to be involved (THz frequency vibrations). There is only a weak coupling between vibrations and the $D_2/^4He$ transition; so we require other (strongly coupled) nuclear transitions for fractionation. The picture in this case is one where the highly excited oscillator is weakly coupled to the $D_2/^4He$ transition, strongly coupled to internal transitions in the deuterons, and coupled to the host metal electrons as a source of off-resonant loss. In the model, the $D_2/^4He$ transition occurs with a single phonon exchange with the large nuclear energy quantum transferred to other more strongly coupled transitions and subdivided (many nuclear excitations for a single $D_2/^4He$ de-excitation), and subsequently fractionated to optical phonons.

While this general approach in our view is consistent with experiments, a major issue is that it is hard to be certain at this point since excess heat is basically ‘silent’ (in that there is nothing energetic emitted in the primary reaction). Because of this, our focus more recently has been on experiments in which energy exchange occurs between vibrations and nuclei under conditions where it is possible to get more information. An example of this is X-ray emission in the Karabut experiment, in which collimated X-rays near 1.5 keV are observed from the cathode during the discharge, and with bursts of collimated X-rays detected for roughly a millisecond after the turn-off of the discharge[36]. According to our interpretation of this experiment, energy is transferred from a strongly excited acoustic vibrational mode (with up-conversion) to produce excitation of the 1565 eV excited state in ^{201}Hg (which we imagine is present inadvertently as an impurity on the surface). Collimated X-ray emission in this picture is a consequence of a phased array effect due to coherence among the transitions produced by the up-conversion[3,32].

Appendix 1. Issues raised by reviewers

Many interesting questions about the interaction and mechanisms proposed in this article were put forth by the reviewers. It seems appropriate to present some of these questions, and to discuss them further.

Background electron density

A low background electron density (below 0.05–0.06 e/Å^3) is required for molecular dideuterium formation in the vicinity of a vacancy; otherwise significant occupation of anti-bonding orbitals occurs, pushing the deuterium atoms apart. However, a higher electron density would produce improved screening, leading to a larger tunnelling probability. Consequently, there is a trade-off. We agree that such a trade-off occurs here. The thought is that the coherent processes under consideration require a reasonably stable D_2 configuration, which is available near an O-site in the vicinity of a monovacancy, as long as the other O-sites around the vacancy are occupied. Because of this requirement, we have to settle for the low background electron density if we rely on coherent processes.

Contribution of loss

A reviewer posed a number questions about the role of loss in connection with fractionation, suggesting energy loss to the bath could under some conditions dominate, and if so perhaps could be optimized. In response, historically most of the analysis has focused on an idealized limit of the model in which the loss is taken to be infinitely fast where it can occur. Perhaps counter to what would be expected based on intuition, no energy makes it to the bath in this limit. Only recently have we expanded our analysis to include more realistic loss models with finite loss that is energy-dependent. In such models some of the energy goes into the bath. We see that the rate of coherent energy exchange under conditions of fractionation is sensitive to the details of the strength and energy distribution of the bath. We note that the primary function of the loss is to break the destructive interference present in the lossless version of the model; as such, we expect efficient coherent energy exchange to the oscillator with low bath loss. This is an issue of interest in our current studies. Also, we would expect that if loss to the bath were dominant, the overall process should become mostly incoherent, in which case there would be no acceleration of the rate that can occur in a coherent quantum process.

Energetic phonons

A reviewer wondered about how there could be an energetic mega-electron volt-level phonon. The simple answer is that such a thing is not possible. However, it may be worthwhile to think about the associated issues. We are familiar with the picture normally studied in textbooks and in the literature, in which the vibrational, electronic and nuclear degrees of freedom are reasonably well separated in a solid. As such, we are able to consider the different modes and energy levels of the vibrational system alone. In this case we end up with a large number of modes which are quantized individually to give approximately simple harmonic oscillators. The creation or destruction of a single phonon in this picture comes then with the exchange of one (low energy) phonon quantum of energy.

If the nuclear system is treated relativistically, there is a very strong coupling present between the vibrational and internal nuclear degrees of freedom. We might ask if this is so, then why do we treat them as nearly independent degrees of freedom in textbooks. The answer is that there exists a unitary transformation that eliminates this very strong first-order coupling. Under conditions where this unitary transformation is useful, the vibrational and nuclear degrees of freedom are nearly independent.

Within mainstream science the view is that the unitary transformation which eliminates the strong first-order coupling is universal and can always be applied. We have noticed that the spin-boson model is similar algebraically, and there is a similar unitary transformation which eliminates the strong first-order coupling. One can make use of this transformation to derive the coherent energy exchange rate in the multi-photon regime of the model when there is no loss. However, in the presence of the different loss models that we have worked with, we find a strongly enhanced rate for coherent energy exchange under conditions of fractionation, which is due to the partial or total removal of destructive interference. The spin-boson models in the presence of loss mechanisms which spoil the destructive interference behave as if the strong first-order coupling is still present. Similarly, we expect that when the analogous destructive interference is spoiled in the version of the solid state model with a relativistic description of the nuclei, there will be a similar enhanced rate for coherent energy exchange under conditions of fractionation. This appears to be the case in the models we have analysed.

Consequently, the picture that emerges is one that can behave differently than what we are used to. If we focus on the ability of the coupled vibrational and nuclear system to coherently exchange energy under conditions of fractionation, then when the associated destructive interference remains there is no fractionation and the lattice acts in the way discussed in textbooks. When the destructive interference is removed then it acts differently. From our analysis we require a highly excited vibrational mode and loss for the destructive interference to be removed. In this case we are dealing with two strongly coupled quantum systems, in which case we should not be surprised to see new effects. The creation or destruction of a phonon in this case can be associated with the exchange of a much larger amount of energy, since the states of the strongly coupled system now are a mix of the vibrational and nuclear degrees of freedom. According to the models we have studied, there is the possibility that a large energy quantum can be exchanged in this case with the creation or destruction of a single phonon, because of a mixing of the vibrational and nuclear degrees of freedom. To refer to this as an energetic mega-electron volt-level phonon in our view would be inappropriate.

Energy localization

A reviewer proposed that the fractionated energy would end up localized in the vicinity of the nucleus where the transition energy originated. In response, we note that the models which show coherent energy exchange under conditions of fractionation describe the coupling of two-level systems with a single oscillator (in the presence of loss). Consequently, the energy exchange is with the vibration of a single delocalized vibrational model. Energy localization as proposed by the reviewer would require the coupling of energy into a large number of different vibrational modes.

Time required for fractionation

A reviewer was interested in whether the energy from fractionation was distributed instantaneously, or sequentially over an extended timescale. Once again the question is interesting, and worth some thought. We have studied models that correspond to different schemes over the years. For example, in a model in which transitions are available to long-lived nuclear states in the hundred kilo-electron volt regime, then a 24 MeV transition can be rapidly followed by many nuclear excitations (which we have referred to as a subdivision of the energy), and subsequently fractionated over a longer timescale. This picture may be relevant when the overall process is mediated by a highly excited acoustic mode, in which case one would expect excitation of the host metal nuclei (and the possibility of subsequent nuclear disintegration).

The results in the two-laser experiment in PdD suggest that in this case optical phonon modes are involved, and the vibrational motion is dominated by the deuterium atoms. This leads to a different picture in which internal transitions in the deuteron produce fractionation. In this case there are no long-lived intermediate states, and so no subdivision of the large 24 MeV quantum; instead we would require fractionation directly into optical phonons.

Models we have studied in this case are based on all of the relevant $D_2/^4He$ transitions coupling to a common, highly excited vibrational mode. The rate of fractionation without subdivision then has to match the energy release rate. For example, if the system produces excess heat at the 1 W level, then there are 2.6×10^{11} reactions/sec and it must take 3.8×10^{-12} sec for each of the large 24 MeV quanta to be fractionated. If the optical phonon mode has an energy of 36 MeV, then the average time associated for the net transfer of a single phonon in connection with fractionation must be 5.7×10^{-21} sec. These numbers are consistent with the models we have studied over the years.

D_2 and excited states of ^4He

A reviewer had commented about the possibility of excited state of ^4He being involved. There is discussion in the literature in the case of the fusion of heavier nuclei that an excited compound state can form, which subsequently sheds its energy via all accessible decay processes. Huizenga argued that conventional deuteron–deuteron fusion reactions similarly go through an excited $^4He^*$ Bohr state[37], but this is not reflected in the theoretical models in the literature[38]. The problem in connection with coherent models is that the excited states of the alpha particle in the relevant energy range are very unstable, which makes them of little use for us. We have studied direct $D_2/^4He$ transitions mediated by phonon exchange[34], where both the initial D_2 and final ^4He states have good stability.

Internal transitions in the deuteron

A reviewer had argued that there are no accessible transitions in the deuteron, so that it could not be involved in fractionation. In response we note that there are two different kinds of transitions that might be relevant. We studied a model for transitions in the deuteron between the ground state and continuum n + p states[33], which leads to a coupling matrix element that seems to us to be too small for the (interesting) anomalous regime of the model discussed earlier[32], although the selection rules appear to be consistent with the magnetic field effects reported recently.

We expect much stronger transitions to negative energy states, with the same selection rules, but these transitions have not been similarly analysed yet. A new version of the model of Hagelstein and Chaudhary[32] is required for the interaction of a highly excited vibrational model with transitions involving negative energy states, but we expect qualitatively similar results. Note that there is no difficulty in proposing models in which a vibrational degree of freedom couples with a negative energy transition; an issue is that there cannot be the possibility of a nucleus ending up in a negative energy state as a result of a physical process.

Mathematical model and physical picture

One of the reviewers argued that in the discussion above, the focus has been on mathematical models and issues, and the reviewer wanted some discussion of a corresponding physical picture. In general this seems to be worthy of further discussion.

Perhaps the place to start is with nuclei in a lattice with a highly excited vibrational mode. In a textbook version of the model we can envision the nuclei as hard particles with an internal structure too stiff to respond to the acceleration of the 'slow' lattice vibrations. In our models there are internal nuclear degrees of freedom which under some conditions can be important; this suggests a physical picture in which the nuclear response needs to be included. Thinking of a nucleus as a water balloon with internal sloshing associated with acceleration is helpful only qualitatively, since the response is still very stiff. Unfortunately, the relativistic coupling of the centre of mass motion with the internal spin degrees of freedom is not easy to visualize under any circumstances, largely because we do not have a relevant good microscopic model for spin that we can visualize in the first place.

Another place where a physical picture would be useful for visualization is in connection with the energy exchange. This has been a headache for many years, since the pictures that result seem not to be intuitive. For example, we might consider energy exchange in the multi-photon regime of the spin-boson model in the absence of loss, which is well studied in the literature. We have gone to some effort to extract out the two-level system dynamics and the oscillator dynamics from numerically exact global solutions under conditions where coherent energy exchange occurs. In these models the oscillator gains energy slowly, and then loses energy slowly, and excitation goes back and forth between the two degrees of freedom. The same is seen in the two-level system dynamics. The indirect coupling that produces the effect in this case is a weak and subtle effect, which does its business with little fanfare. One can watch the dynamics, see the results which are at best non-intuitive and a little disconcerting, and understand them only because one knows from the mathematics that there is a weak indirect coupling that allows energy exchange between the two mismatched degrees of freedom.

When the destructive interference is spoiled, so that the indirect coupling under conditions of fractionation becomes greatly accelerated, there is little change in the basic picture that emerges above. The oscillator still exchanges energy slowly (although now the fractionation is much greater); the two-level system dynamics is similar, as one can see that transitions occur over a long timescale. Once again there are no fireworks, since the underlying mechanism at work does its business as a result of a large number of mundane interactions. In the end it remains the case that the way energy is exchanged is through a large number of individual interactions where one phonon exchange is coupled to the excitation or de-excitation of a two-level system, a process which continues until the system loses coherence.

The physical picture that results in the end for excess heat production in PdD with a high excited optical phonon mode is simple and non-intuitive. One arranges for molecular D_2 near the O-sites of vacancies; the more the better. In the two-laser experiment, the initial excitation of optical phonons at the Γ-point is produced over roughly a square millimetre of the surface (subsequently expanding to a larger area), where vacancies and molecular D_2 are present. In optical phonon vibrations at the Γ-point, all of the bulk PdD deuterons near the surface involved simply oscillate back and forth in phase less than 0.1 Å. Since the vibrational excitation in the experiment that is effective is compressional, there is a weak squeezing effect that brings the two deuterons in the molecular D_2 states slightly closer together. In the model there is strong coupling between the vibrational motion and the internal degrees of freedom of the bulk deuterons, probably involving transitions to negative energy states, which allows for fractionation but otherwise does not do much else. So as we imagine ourselves watching with microscopic eyes, we see molecular D_2 being converted in place to ^{4}He with the energy fractionated and going into the optical phonon vibrations. In these models, there are no fireworks at all; the transitions occur with no local energy deposition, and the large quanta are fractionated as a consequence of the coupling of the vibrations with the internal nuclear degrees of freedom of the deuterons in the presence of loss. Loss is present since there are conduction electrons that can be promoted, bound electrons that can be ionized, and nuclei that can be excited or disintegrated (all of which we expect to occur at low levels, since the dominant coherent process is so much faster). In our mind's eye we visualize phonon gain as a consequence of the fractionation of the nuclear quanta in the optical phonon modes, where the pushing comes from the equivalent force on the deuterons that follows from the fractionation dynamics. The surface phonon laser is driven by the nuclear transitions, but due to the very short (picosecond) lifetime of the optical phonons, and the low group velocity of the Γ-point vibrations, the phonon laser energy remains localized and simply thermalizes.

Probably we could see the surface vacancies using a thermal desorption measurement, and perhaps we might be able to see the molecular D_2 in a suitable magic-angle NMR experiment. We could imagine a Raman measurement would show us that optical phonon modes were highly excited in connection with excess heat production. We could also imagine that ^{4}He accumulates in the vacancies and limits the rate at which excess heat is generated, until the helium leaves the monovacancy traps and diffuses to the surface, where it could be detected in the gas phase.

Quantification of the model

One of the reviewers remarked that readers would be unable to extract physical meaning from the various models in the absence of a quantitative estimate for all of the coupling terms and interactions under discussion. In response we note that the issue of quantitative analysis has been important in our work from the beginning, and over the years we have developed a large number of individual estimates. The most important parameters must be the coupling matrix elements associated with the coupling of the vibrational and internal nuclear degrees of freedom. Considerable effort has gone into the computation of the $D_2/^4$He matrix element, as described in ref. 34, where explicit numbers are given. In our view this matrix element is sufficiently large to account for experiments; with best estimates for screening and model parameters we obtain consistency with experimental rates for excess heat production. A model for the phonon exchange matrix element has been studied in the case of the deuteron as well[33]. This time we are of the opinion that the matrix element is too small to fractionate a 24 MeV quantum, based on our most recent modelling[32]. The problem in our view is that we focused on the d/n + p transition, which is weak; instead we should probably have considered coupling from the ground state to negative energy states, which would be much stronger.

To make use of the new models systematically for the anomalies of interest in condensed matter nuclear science more generally, we may have to develop a new computational capability. For example, we would like to compute phonon exchange matrix elements for coupling to negative energy states, as well as to high-spin states in Pd and Ni. Also of interest is the phonon exchange matrix element for excitation to the 1565 eV transition in ^{201}Hg. We hope to make progress in this direction in the future.

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