Directional X-ray and gamma emission in experiments in condensed matter nuclear science

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The absence of energetic particles commensurate with the energy produced in the Fleischmann–Pons experiment hinders our ability to sort out the microscopic physics involved. Models that we have studied for excess heat are based on the fractionation of the large nuclear quantum to a large number of much smaller quanta. These models predict that it should also be possible to up-convert vibrational quanta to produce nuclear excitation. Such a mechanism could produce collimated X-ray and gamma emission. Collimated X-ray emission near 1.5 keV has been reported by Karabut. Other examples of collimated X-ray and gamma emission have been described in the literature.

Keywords: Cold fusion, excess heat, nuclear excitation, X-ray and gamma emission.

Introduction

EXCESS heat in the Fleischmann–Pons experiment\textsuperscript{1,2} is thought to have a nuclear origin; however, energetic nuclear products are not present in amounts commensurate with the energy produced\textsuperscript{3–5}. This makes it clear that whatever process is involved, it must work differently than conventional nuclear reaction processes. The absence of commensurate energetic products also greatly hinders our ability to clarify how the associated microscopic reaction mechanism works. It almost seems as if there is a curtain up which prevents us from seeing what goes on inside the experiment. It is this lack of a conventional energetic nuclear signature which hinders progress on clarifying theoretical issues, and which also leads to skepticism within the physics community (who simply cannot accept that the energy produced can have a nuclear origin without the presence of commensurate energetic nuclear emissions).

In our view, this absence of commensurate energetic particles is the most important feature of the process which must be addressed theoretically\textsuperscript{7}. For us, it signals clearly that it must be possible to fractionate a large megaelectron volt-scale nuclear quantum into a very large number of much smaller electron volt-scale quanta. No alternative appears to be viable.

A substantial effort was devoted over many years searching for models capable of coherent energy exchange under conditions where a large quantum is fractionated into a great many small quanta. It is known that coherent energy exchange can occur in the multiphoton regime of the spin-boson model, in which the large transition energy of (identical) two-level systems can be converted into a modest (odd) number of oscillator quanta. The rate for energy conversion in this system is limited by a destructive interference effect (where contributions to the weak indirect coupling matrix element from different pathways destructively interfere). We found that when an appropriate loss mechanism is introduced, the coherent energy exchange rate is increased by orders of magnitude, and the system is able to exchange energy coherently, even when the oscillator energy is a very small fraction of the two-level system energy. We developed simple models for this effect\textsuperscript{8–11}. A version of this kind of model has been adapted to describe excess heat production in the Fleischmann–Pons experiment\textsuperscript{12}. The mechanism considered is capable of fractionating the large nuclear quantum into much smaller electron volt-scale quanta coherently; energetic nuclear products are suppressed naturally within the theory because the coherent (and nuclear radiation-free) process can be many orders of magnitude faster than the incoherent processes that produce commensurate amounts of nuclear radiation.

This leads to a situation in which there can be consistency between experiment and theory, with no commensurate energetic nuclear radiation present in either. Unfortunately this is insufficient, as the absence of commensurate energetic products in the model is not enough to provide strong confidence that the model is correct. We really need a new experiment in which coherent energy exchange between highly mismatched systems leads to the emission of an unmistakable energetic product. If we really can convert a large nuclear quantum into many small vibrational quanta, then we would like to have an experiment that demonstrates the conversion of a large number of vibrational quanta to produce excitation in a nucleus, under conditions where we can prove that the nucleus has been excited from its (unambiguous) decay products.

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Design issues for an experiment

It seems worthwhile to consider briefly design issues for an experiment to demonstrate this kind of up-conversion. According to the models, there is no problem with either the up-conversion or down-conversion of vibrational energy from a (lossy) highly excited vibrational mode to a nuclear transition; however, it becomes easier when the number of small quanta \( \Delta n \) is less. For example, we would expect for a conventional nonlinear interaction for a 11th-order process to be slower than for a third-order process; correspondingly, we would expect fractionation with \( \Delta n \) on the order of \( 10^6 \) to be faster than fractionation with \( \Delta n \) on the order of \( 10^9 \). This expectation is consistent with the maximum transition rate computed from the model\(^{11,12}\).

As a result, if we would like to up-convert a large number of vibrational quanta to produce nuclear excitation, then we are motivated to find the lowest energy nuclear transition (consistent with the ground state being stable to make the experiment easier to perform; and consistent with the transition involving the ground state, as greatly favoured by theory). This leads us to a consideration of the 1565 eV transition in \(^{201}\)Hg as most interesting (Table 1). \(^{17}\)

A question arose in the review of this paper as to why electronic transitions are not included in the discussion. We note that during the initial design effort (and subsequent analysis effort) electronic transitions were included as candidate transitions on equal footing to nuclear transitions. A major difficulty is that the electronic states have very short lifetimes in the kiloelectron volt regime, which makes it difficult to develop massive up-conversion; it requires that coherence be maintained long enough to complete. Another issue is that the coupling that we estimated between electronic excitation and lattice vibrations is weak for inner-shell transitions. Ultimately the model suggested that nuclear transitions (mediated by the relativistic interaction\(^{17}\)) should be much more effective in fractionation or inverse fractionation.

In order to minimize \( \Delta n \), we would like to work with the highest vibrational frequencies possible for a macroscopic sample, which are in the terahertz range. We know from theory that we would also like a large number of nuclei (\( N_0 \)) moving coherently in order to reach the threshold condition for the onset of fractionation. In an important limit of one version of the model, the ratio \( N_0/\Delta n^2 \) needs to be maximized to reach the threshold for the onset of fractionation\(^{15}\). Since in the terahertz regime it is difficult to establish coherence over a sizable region (without gain present), backing off to a lower frequency in a sample without gain makes sense. Since \( N_0 \sim \Delta n^2 \), we probably want to work at the highest frequency in which the vibrations are coherent over the whole sample, which argues for 0.1–1 GHz vibrations in a coin-sized sample.

In our initial design considerations, we contemplated working with samples in which the Hg content was maximized. Eventually we understood that other transitions could accomplish the fractionation, and that the excitation could be transferred to the 1565 eV transition in \(^{201}\)Hg. As a result, Hg only needs to be on the surface. If the 1565 eV transition is excited, we expect electron emission by internal conversion decay to be dominant; however, there is also a weak radiative decay of this transition\(^{15}\).

We recognized that if the up-conversion is from a vibrational mode that is uniform across the surface of the (coin-shaped) sample, then we might expect that the X-ray emission should be directional due to a phased antenna array directional radiation effect. Models for the line shape suggest that the emission line should be quite broad\(^{14,16}\).

### Karabut’s collimated X-ray emission near 1.5 keV

In light of the discussion above, we draw attention to observations of directional X-ray emission in Karabut’s high current density glow discharge experiments. Karabut first observed the effect back in 2002 (ref. 17), and reported many subsequent studies of the effect over the years\(^{18–20}\). He detected the X-rays with a scintillator, with a thermal luminescent detector, imaged on film with an X-ray pinhole camera, and spectrally resolved on film using a bent mica spectrometer. Karabut’s collimated X-ray emission appears as a diffuse broad feature in the vicinity of 1.5 keV in data taken with the discharge on. Very bright flashes of X-ray emission are seen over the course of a millisecond timescale after the discharge is turned off suddenly. In the latter case, the film is damaged due to the intensity of the X-rays, and the spatial localization can be on the 1 mm scale (in the case of a centimetre-sized cathode). The X-rays associated with the bright flashes seem to be very narrow in energy, but occur at random energies between about 500 eV and 5 keV. Some of these appear as streaks on the film, suggesting a searchlight emission effect from the cathode surface\(^ {21}\).

This directional X-ray emission is a reproducible effect in Karabut’s experiments, and depends only weakly on

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**Table 1.** Low-energy nuclear transitions from the ground state of stable nuclei, from the BNL on-line NUDAT2 table

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Excited state energy (keV)</th>
<th>Half-life</th>
<th>Multipolarity</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{201})Hg</td>
<td>1.5648</td>
<td>81 ns</td>
<td>M1 + E2</td>
</tr>
<tr>
<td>(^{181})Ta</td>
<td>6.240</td>
<td>6.05 µs</td>
<td>E1</td>
</tr>
<tr>
<td>(^{169})Tm</td>
<td>8.41017</td>
<td>4.09 ns</td>
<td>M1 + E2</td>
</tr>
<tr>
<td>(^{83})Kr</td>
<td>9.4051</td>
<td>154.4 ns</td>
<td>M1 + E2</td>
</tr>
<tr>
<td>(^{187})Os</td>
<td>9.75</td>
<td>2.38 ns</td>
<td>M1(3E2)</td>
</tr>
<tr>
<td>(^{73})Ge</td>
<td>13.2845</td>
<td>2.92 µs</td>
<td>E2</td>
</tr>
<tr>
<td>(^{57})Fe</td>
<td>14.4129</td>
<td>98.3 ns</td>
<td>M1 + E2</td>
</tr>
</tbody>
</table>
the cathode metal used (a wide variety of metals between Al and Pt), and on which gas discharge is used (a moderate variety of gases, including H₂, D₂, Ar, Kr and Xe). Karabut reported that it was possible to maximize the X-ray emission so that the radiated X-ray power was on the order of 20% of the input power to the discharge. We have proposed that Karabut’s collimated X-ray emission is a consequence of the up-conversion of vibrational energy to produce excitation in ⁴⁰⁰K₃, which may be present in small amounts on the surface as an impurity. The supposition is that the number of Hg atoms present on the surface is very low (on the order of 10⁻¹⁰⁻¹) (ref. 16), which is consistent with a ubiquitous level of Hg contamination, keeping in mind that all accessible contaminants will be sputtered onto the cathode surface in a glow discharge.

Proposed interpretation

Highly directional X-ray emission near 1.5 keV from the surface of a cathode in a glow discharge experiment seems to be difficult to account for theoretically based on conventional approaches. To account for the directionality and intensity of the emission, Karabut has proposed that an X-ray laser is responsible (although the possibility of up-converting vibrational quanta was noted). From our perspective, the development of an X-ray laser under these conditions seems to be unlikely, especially when bright flashes are seen on the order of a millisecond after the discharge is turned off. For example, an excited atomic electron in the kiloelectron volt regime decays rapidly (on a femtosecond timescale).

Plasma decay and afterglow have been studied under roughly comparable conditions in other experiments in Ar and Ne, with the result that the plasma decay takes a few tens of microseconds (but the afterglow due to recombination can last longer). From the current and voltage traces after the discharge is turned off, one sees that there is no subsequent significant electrical input into the gas in Karabut’s experiment when the bright flashes are seen. The energetic electrons distribution in the plasma loses its energy within about 20 μs (ref. 29), so that there are no energetic electrons around to cause kiloelectron volt excitation at the time of the flashes. In a glow discharge, the electric field accelerates electrons towards the anode, where the collimated X-ray emission is observed originating from the cathode.

Note that aside from the 1565 eV transition in ¹⁷¹Hg, there are no nuclear transitions from the ground state of any other stable nucleus in the spectral regime under discussion.

Motivated by the theoretical ideas outlined above, we view Karabut’s high current glow discharge as a source for exciting acoustic vibrations in the 50–200 MHz range in metal cathodes (which are 10–50 μm thick). The reason for this is that we worked with a version of Karabut’s glow discharge in our lab at MIT in the mid-1990s, when the issue of large amplitude voltage spikes came up. Karabut observed very short (nanosecond, or sub-nanosecond) voltage spikes measured in excess of 50 kV in discharges run below 2 kV. The analogous voltage spikes in our glow discharge did not exceed 10 kV, which we attributed to different drive electronics (in this regard see ref. 32 which contains some documentation of the circuit used by Karabut).
Collimated X-rays in the experiments of Kornilova et al.

We also draw attention to experiments by Kornilova, Vysotskii, and co-workers, who reported directional X-ray emission from a steel plate next to a high pressure water jet\textsuperscript{35,36}. As is worth emphasizing, collimated X-ray emission is not an effect that would be expected unless either an X-ray laser has been produced, or a collimated electron beam was nearby, or else phase coherence was present among the atomic or nuclear states. The authors attribute the effect to conventional physics based on shock wave generation, followed by atomic excitation. Our interpretation of the experiment is different: we consider the water jet to be a strong broadband acoustic noise source, and the 3 mm thick steel plate to be an acoustic resonator with a fundamental compressional mode near 1 MHz

$$\omega_0 = \frac{c \pi}{d} = 6.1 \times 10^5 \text{cm sec}^{-1} \pi \text{sec} 3 \text{mm} \approx 2 \pi \times 10^{11} \text{rad sec}^{-1}.$$ 

with higher-order modes at higher frequencies. If the mechanism is similar to what we propose for Karabut’s experiment, then strong excitation of the fundamental may result in up-conversion of the vibrational excitation by a very large factor ($O(10^{11})$) to produce phase coherent nuclear excitation, resulting in collimated X-ray emission.

In these papers\textsuperscript{35,36} appear results based on X-ray film, and also on an AMTEK X-123 spectrometer. We consider the X-ray film to be more reliable in this case, and are concerned about the measurements reported with the X-123. The issue in this case is that there is a Be window in the spectrometer that prevents X-rays below a threshold energy (near 900 eV) to make it to the silicon detector. Hence, if the detector response is due to X-rays, then one might expect to see a cut-off in the data. There is no indication of this in the data presented, which leads to a concern that the instrument is responding to something other than X-rays (perhaps ultrasound, or possibly charge).

The vibrating copper foil experiment at SRI

We were interested in developing an experiment to clarify the mechanism of collimated 1.5 keV X-ray emission in Karabut’s experiment. If the theoretical model is right, then we should be able to generate X-ray emission by vibrating a thin metal sample with Hg on the surface. An experiment was set up at SRI seeking to test this\textsuperscript{37}. A thin (73 μm thick) copper foil was selected for the tests; copper was favoured for this since mercury is known to bind well to the surface (before diffusing into the bulk). Tests were done initially to verify from current and voltage measurements that excitation of the vibrational modes took place through capacitive coupling of a thick driver with the foil; in some cases a response can be seen in the data.

Measurements were done to look for charge emission from the foil, since the decay of the 1565 eV level in \textsuperscript{201}Hg is predominantly through internal conversion (in which an electron is emitted). An electrometer was used to look for charge, and strong reproducible signals were seen at frequencies which match the expected compressional and transverse frequencies (near 15 MHz drive frequency for the second transverse mode, and near 17 MHz drive frequency for the first compressional mode). The charge emission was found to be uncorrelated with surface Hg; it was negative in sign and was not accompanied by optical emission. This charge emission effect is anomalous; the conjecture currently being contemplated is that it is due to the up-conversion of the vibrational quanta to promote conduction electrons which make it out of the metal to form negative O\textsuperscript{2-} ions in the air.

An AMPEK X-123 detector was used to monitor X-ray emission from the foil, under conditions with and without surface Hg. Strong signals were observed reproducibly below 2 keV, seemingly correlated with surface Hg (as reported at the colloquium at MIT in March 2014). However, these signals appeared both above and below the Be window cut-off energy near 900 eV, which means that they are not due to X-rays in the region where the detector is sensitive. In some experiments where no Hg was present and the detector was well-grounded, it was possible to obtain clean spectra that had no counts above 900 eV, and a very low number of counts down to pile-up. Similar experiments done with surface Hg gave counts above pile-up, and in some cases above 1 keV. One possibility is that the detector responds to charge emission from the surface when not well-grounded, and that charge is emitted from the foil when the vibrational resonances are strongly excited as discussed above. A concern that has not yet been pursued is that the detector responds to ultrasound.

Rhodes’ group X-ray laser experiments

In the models that we have explored for the anomalies in condensed matter nuclear science, there are two distinct parts that are new: a mechanism for fractionation (and inverse fractionation), and a relativistic Hamiltonian that describes coupling between vibrations and internal nuclear degrees of freedom. It would be important to pursue each of these independently in order to be sure that they correspond to experiment, which provides us with motivation to consider the possibility that fractionation, or inverse fractionation, might occur in the up-conversion of electromagnetic quanta (photons) under some conditions. Some thought has gone into the possibility of setting up a ‘conventional’ experiment in a laser cavity with appropriate
loss to check whether enhanced up-conversion through inverse fractionation might occur.

However, more than a decade ago, an X-ray laser experiment was reported by Rhodes and co-workers \(^{38,39}\) that may be relevant in this discussion. The scaling laws which govern laser amplifiers dictate that higher power density is required in order to develop a population inversion through collisional excitation or ionization. As a result, when the possibility of confining an intense optical laser pulse in a narrow plasma channel arose, Rhodes and colleagues focused their efforts on utilizing the effect to develop an X-ray laser. Although there have been variants on the theme over the years, the basic idea which the group focused on was the possibility of a collectively enhanced inner-shell ionization effect that would create a population inversion on a self-terminating transitions with a very short wavelength. In early experiments spectra were recorded that were interpreted by the experimental group as consistent with much more highly ionized ions that would be expected through tunnel ionization. In 2002, the group reported the observation of strong directional emission below 3 Å in Xe clusters, and in the following years they claimed to have produced a saturated X-ray laser.

These experimental results have met with some skepticism, as they are in a sense too good to be true. Usually a claim of lasing in the X-ray regime is made with a demonstration of exponentiation with amplifier length, which in this case is missing. Also, since the beginning experimental X-ray laser work was closely connected with detailed simulations of the laser kinetics (as the associated experiments were so expensive, theory was important in improving the chances of success). However, there is no adequate detailed theory for the Rhodes X-ray laser. Simulations have been carried out at NRL\(^{40,41}\), which show that it is possible to develop sufficient gain on transitions in the vicinity of those observed; however, these simulations are based on an assumption that either X-rays or electrons are present which preferentially ionize inner-shell electrons. There is no evidence for the corresponding X-rays in the experimental spectrum, and there are no systematic calculations showing preferential inner-shell collisional ionization for the ions involved.

Our group was tasked by Darpa to attempt the development of a theoretical model for the Rhodes’ group X-ray laser. Over the course of about two years, a number of models were constructed to study enhanced inner-shell collisional ionization as proposed by Rhodes’ group. The results of this work have not yet been published; however, an enhanced effect was found which involved outer-shell ionization over inner-shell ionization, and a mechanism was found which at sufficiently high intensity would result in enhanced tunnelling of the outer-shell electrons by the optical laser.

Ultimately we came to the realization that it might be possible to account for the experimental results through an enhanced multi-photon excitation effect in which the optical photons interacted with strongly coupled electronic transitions in the highly stripped ions in the presence of (photoionization) loss. In a sense this system can be described by models that are like the lossy spin-boson model that we have studied, and which show inverse fractionation. We developed an exploratory model to describe inverse fractionation under these conditions, and the results of the model agree well in the case of the recent experimental results in Ne-like Kr\(^{42}\). A preliminary account of the modelling results in Kr and Xe clusters is given in ref. 43. We plan on completing a manuscript with a detailed discussion of the new models in the coming months.

Discussion

Collimated X-ray emission near 1.5 keV in Karabut’s glow discharge experiments in our view is of fundamental importance in sorting out the new physics involved in excess heat in the Fleischmann–Pons experiment. The difficulty in understanding the excess heat effect is that we are unable to observe the final products of the reaction directly as energetic particles (since there are no commensurate energetic particles present). If the large nuclear quantum is being fractionated in these experiments, we could have consistency with theory, but with no clear positive proof. On the other hand, the models indicate that it should be possible to go the other way, and up-convert a large number of low-energy vibrational quanta to produce nuclear excitation. If so, then we should be able to make it happen, observe the radiation produced, and study it systematically. Karabut’s collimated X-rays in our interpretation show that this is possible. Added support for this point of view comes from Gozzi’s collimated gamma rays, Karabut’s collimated penetrating X-rays, and the directional X-rays seen in the experiments of Kornilova, Vysotskii and co-workers. The experiment begun at SRI was intended to clarify Karabut’s collimated X-rays; however, within the limited time available for experimentation it was not possible to complete what we had hoped to do. The charge emission effect observed appears to be a reproducible anomaly, and which may be a precursor to the 1.5 keV X-ray emission if indeed the charge emission is due to up-conversion of the vibrational quanta.

We recognize the importance of demonstrating up-conversion of low-energy quanta in experiments outside of condensed matter nuclear science, since the associated mechanism is general, and not limited to the vibrational/nuclear system. We have for some time been interested in the up-conversion of photons through an inverse fractionation mechanism, and in the possibility of a demonstration in a conventional laser cavity setting. However, it seems to us that the stunning experimental X-ray results reported by Rhodes’ group over the past decade and more have no other explanation.
Appendix 1. Absence of fractionation effect in perturbation theory.

A reviewer has provided an objection that the massive up-conversion effect under discussion is simply impossible because it is a high-order effect with a very large number of intermediate virtual states. For example, if we think of $g$ as the ratio of the interaction matrix element to the transition energy, then under normal circumstances we would expect a perturbation theory calculation to result in a (coherent) rate that scales roughly as $g^4$, with $g$ less than unity. In the models that we have studied, we see exactly this effect, which leads to astronomically small indirect coupling matrix elements when the coupling is too weak.

However, when there are many (identical) two-level systems, the interaction matrix element is increased with Dicke coherence factors, so that the ratio $g$ greatly exceeds unity (as long as the oscillator is highly excited). In this regime perturbation theory becomes unhelpful, as well as our intuition that derives from perturbation theory. This regime occurs in the spin-boson model (where $g$ can be much larger than unity), where non-perturbative methods are required, and where a small coherent energy transfer rate can result for modest $\Delta n$. As mentioned in the introduction, when the system is augmented with an appropriate loss model one finds a dramatic increase in the indirect coupling matrix element for coherent energy exchange. Several models of this kind have been analysed with non-perturbative methods.

Appendix 2. Coherent versus incoherent phonons.

A reviewer has argued that massive up-conversion is impossible since the ultrasound excitation in both cases is incoherent with random phases. We note that in the models studied so far, coherent energy exchange under conditions of fractionation requires a highly excited oscillator. We would not expect massive up-conversion for thermal excitation, or for a random excitation of many modes. In the case of the Karabut experiment, there is no independent measurement which shows any vibrational mode is highly excited, as no relevant measurements have been carried out. This is one of the reasons that we are interested in the vibrating copper foil experiment at SRI, since in this case we induce vibrations in the megahertz range, and there is the potential for measuring to see that they are present in connection with anomalous emissions.

In the case of the experiments of Kornilova, Vysotskii and co-workers, our interest has been focused on the observation of collimated X-ray emission from a steel plate next to a high-pressure water jet, as measured with X-ray film. In this case we are not aware of ultrasound measurements done on the plate; so we do not know whether there is strong excitation near 1 MHz.

However, ultrasound measurements were reported by Kornilova, Vysotskii and co-workers on the hard plastic of the Koldomasov system, under conditions where the AMTEK X-123 reported X-ray signals. In this case, broad peaks were seen, which the authors reported at 22 and 44 kHz, with a homogenous signal above 50 kHz. The reviewer argues that this provides a counter-example that is inconsistent with thesis presented in the paper (that there are collimated X-rays and no clear sharp resonance).

As mentioned in the text, in our experiments we observed qualitatively similar signals in the experiments at SRI with a similar X-ray detector, and we initially concluded that we had seen X-rays. However, in our datasets, these signals showed up both above and below the cut-off energy for the Be window; so we understood that the detector was not responding to X-rays. It is possible that the detector was responding to charge, or perhaps ultrasound. In control experiments done with rigorous grounding of the detector to the Cu foil, these signals were eliminated, suggesting that in our experiment the detector responded to charge. We note in figure 4 of Kornilova et al. the signals seen by the AMTEK X-123 transition smoothly from low energy (where no transmission through the Be foil is possible below about 900 eV) to high energy (where a broad feature with a peak at 1.2 keV should show the effects of increasingly strong absorption below the peak). Consequently, we suspect that the X-123 signals are not due to X-rays.

The same reviewer argues that the change in the X-ray spectrum reported in the literature which shows the AMTEK X-123 spectrum increasing at higher energy from hard plastic to Fe, and then to Pb on the surface of Fe, is inconsistent with the mechanism proposed in this work. Once again, we suspect that the AMTEK detector in this case is not responding to X-rays. We noticed in the experiments at SRI that the charge emission seemed to occur at lower levels when Hg was on the surface, suggesting a sensitivity to what is on the surface.
17. Karabut, A. B., Research into powerful solid X-ray laser (wave length is 0.8–1.2 nm) with excitation of high current glow discharge ions. In Proceedings of the 11th International Conference on Emerging Nuclear Energy Systems, Albuquerque, New Mexico, USA, 29 September–4 October 2002, pp. 374–381.