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Research Article

Seeking X-rays and Charge Emission from a Copper Foil Driven at MHz Frequencies

F.L. Tanzella, J. Bao and M.C.H. McKubre
SRI International, Menlo Park, CA, USA

P.L. Hagelstein*
Massachusetts Institute of Technology, Cambridge, MA, USA

Abstract

The absence of energetic nuclear particles in the Fleischmann–Pons experiment has been interpreted as indicating that a large nuclear quantum can be down-converted into a great many low energy vibrational quanta. Models that describe this also suggest that low energy vibrational quanta can be up-converted to produce nuclear excitation. Karabut's collimated X-ray emission has been interpreted as being due to the up-conversion of vibrational energy in a small cathode to produce excitation in ^{201}Hg. To test this, we developed a new experiment to vibrate a copper foil with and without surface Hg, and we looked for X-ray emission and charge emission from the surface. Signals were observed in the detectors in both cases; however, the signals in the X-ray detector did not respect the absorption edge of the Be window and are artifacts; and the large current signals associated with charge emission did not charge a capacitor in a simple configuration. We conclude that both are artifacts. The absence of collimated X-ray emission in this case is interpreted as due to the absence of strong low energy nuclear transitions in the copper, and also as ruling out a candidate theoretical model involving up-conversion due to interactions with negative energy transitions. A new interpretation of the Karabut experiment focuses now on lower frequency vibrations in the massive steel cathode holder and vacuum chamber as responsible for the up-conversion, and transfer of the up-converted energy to surface ^{201}Hg to produce the collimated X-ray emission.

Keywords: Charge emission, Collimated X-ray emission, Karabut experiment, Up-conversion, Vibrations

1. Introduction

The most significant claim made by Fleischmann and Pons in 1989 was for their observation of an unexpected excess heat effect in electrochemical experiments where palladium was loaded with deuterium in a D_2O/LiOD electrolyte [1,2]. No commensurate chemical products were found, which motivated Fleischmann to conjecture that the origin of the energy produced must be nuclear. However, one would expect under normal conditions to observe energetic

*Corresponding author. E-mail: plh@mit.edu

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nuclear radiation in amounts commensurate with the energy, and this is not seen in Fleischmann–Pons experiments [3]. For the scientific community, this absence of commensurate nuclear radiation has been sufficient to conclude the experiments must be in error [4]; for us it is a potential advantage to be able to produce clean nuclear energy in large amounts for possible commercial applications.

This situation leaves us with a number of scientific questions. For example, how might it be possible for nuclear energy to be released without a matching amount of nuclear radiation. A question often asked back in 1989 was: where are the neutrons? However, we recognize now that this absence of a commensurate amount of neutrons also extends to protons, alphas, other nuclear fragments, electrons, and gammas. Another important question has to do with what specific reaction mechanism is involved. For example, in a conventional exothermic nuclear reaction one has the possibility of detecting the reaction products as energetic particles. Based on the measurements of the products one normally can understand most everything important about how the reaction works, and in some cases make use of computation for a theoretical understanding. In the Fleischmann–Pons experiment the reaction mechanism cannot be similarly studied, since no commensurate energetic reaction products are created in the first place. ^{4}He has been detected in the off-gas correlated with the energy produced [5–8]; but due to the absence of secondary neutrons we know that the ^{4}He nucleus is born with almost none of the reaction energy [9,10].

All of this in the end leads to a tremendously puzzling situation. We do not know by what mechanism energy is produced in the Fleischmann–Pons experiment, and we cannot make use of normal nuclear diagnostics to study it since the primary reaction mechanism does not produce energetic nuclear products. If a bright theorist managed to deduce the microscopic reaction mechanism responsible, based on what we know it would not be possible to prove it directly (as with conventional nuclear reactions) by performing a Fleischmann–Pons experiment.

A theoretical approach has been developed over many years that is based on the down-conversion of the large nuclear quantum to low energy condensed matter degrees of freedom, such as optical phonons [11–15]. In an experiment which works this way, there should be large amplitude vibrations which could be detected by optical measurements, or using low-energy inelastic neutron scattering. Models developed to describe this effect also predict that vibrational energy should be able to be up-converted to produce nuclear excitation. This raises the possibility that we might be able to study the underlying mechanism responsible for excess heat in the Fleischmann–Pons experiment by developing new experiments that provide vibrational stimulation which is up-converted to produce nuclear excitation, and then detect the subsequent nuclear decay [16].

This line of thought motivated us to seek stable nuclei which have the lowest energy excited states among the stable nuclei, resulting in the identification of candidates such as ^{201}Hg (1565 eV), ^{181}Ta (6240 eV) and ^{181}Tm (8410 eV). For example, the up-conversion of vibrational energy could produce excitation of the 1565 eV transition in ^{201}Hg, which could be diagnosed by looking for electron or X-ray emission. If the nuclei are excited in-phase, it would be possible for the output radiation to be collimated. These considerations focus our attention on the high-current density glow discharge experiments of Karabut [17–22]. In 2002 Karabut reported observations of collimated X-ray emission near 1.5 keV from a variety of different cathodes and a selection of different discharge gases [17]. It was conjectured that strong vibrations in the cathode by voltage spikes in the discharge; that the nuclei in the cathode work together to up-convert the vibrational energy; that the excitation is transferred to a small number of ^{201}Hg nuclei on the surface; and finally that this in-phase excitation of the nuclei results in collimated X-rays through a phased array emission effect [16,23]. The associated line shape is broad, consistent with the broad line shape from the models, suggesting that perhaps Karabut's glow discharge implements the scheme under discussion.

2. The experiment

To make progress on this line of argument, we decided to develop an experiment in which a copper foil could be vibrated over a range of frequencies in the MHz regime, and with different amplitudes; with the possibility of coating a surface with mercury, and looking for electron and X-ray emission from the coated surface [24]. We chose to work with a copper foil since Hg binds well to the surface (and subsequently diffuses slowly into the bulk); and also because Karabut reported positive X-ray emission results with copper cathodes.

There are no measurements of vibrational frequencies excited in Karabut's experiments. From theory we would expect up-conversion to be easier the fewer phonons required in the up-conversion, which focused our attention on the fundamental cathode resonances which were estimated to be in the range of 50–250 MHz. Constraints imposed by the available electronics and supply of copper foils led to the selection of rolled copper foils roughly 73 μm thick for our experiment, with vibrational resonances above 20 MHz.

One surface was capacitively coupled to a thick driver; first through air, and later through PVDF to avoid air breakdown. No DC bias was used so that the frequency of the force on the foil surface was at twice the frequency of the voltage applied to the driver. The applied voltage and frequency was computer controlled through an Agilent 8648A signal generator and amplified with an ENI 603L linear amp. In early experiments we used measurements of the input impedance to try to detect vibrational energy storage and energy loss from the foil. Broad resonances were seen in some cases, which suggested the possibility that vibrational energy might leak into the holder. This motivated us to implement a low-loss cylindrical resonator to hold the foil (Fig. 1).

The 1565 eV excited state of ^{201}Hg decays primarily through internal conversion [25], so we implemented charge detection using a Keithley 617 electrometer, as illustrated in the schematic of Fig. 2. We made use of an Amptek X-123 Si-PIN detector to look for X-ray emission. We were able to measure for charge, or for X-ray emission, but only one at a time.

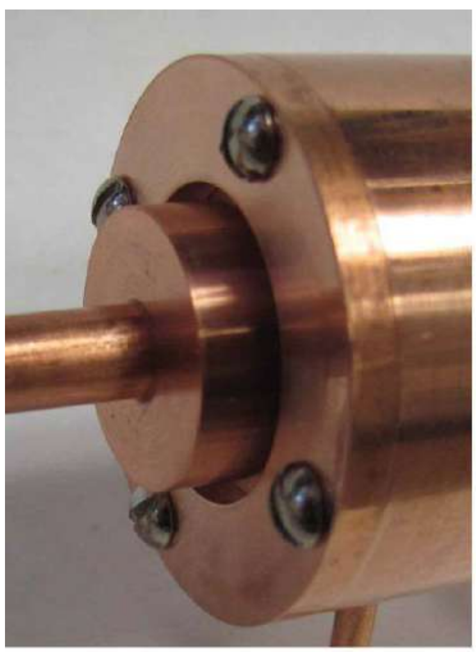

Figure 1. Close-up of driver, foil and resonator.

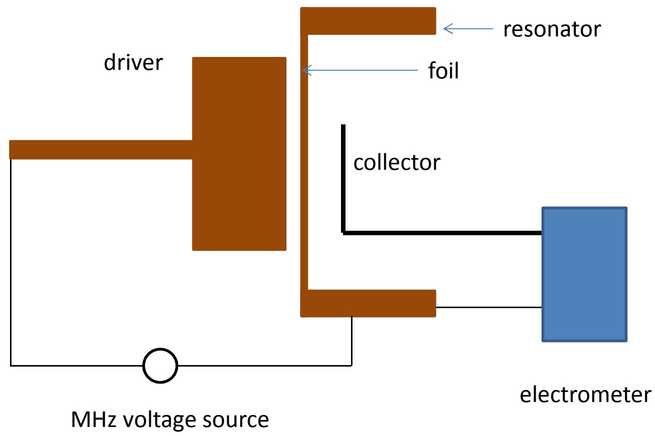

Figure 2. Simplified schematic of foil and resonator, driver, drive circuit and charge detection.

3. Measurement Results and Noise Issues

During the experimental campaign we saw signals on the electrometer that seemed to be consistent with charge emission, and which appeared to be correlated in frequency with some of the expected vibrational resonances; an example is shown in Fig. 3. The sign of the current was negative, consistent with electron emission. A bias on the order of 5 V results in a weak modification of the signal, and a bias near 100 V had a much stronger impact on it. The magnitude of the current signal was somewhat larger with no surface Hg, which suggests that whatever the origin of the effect, it

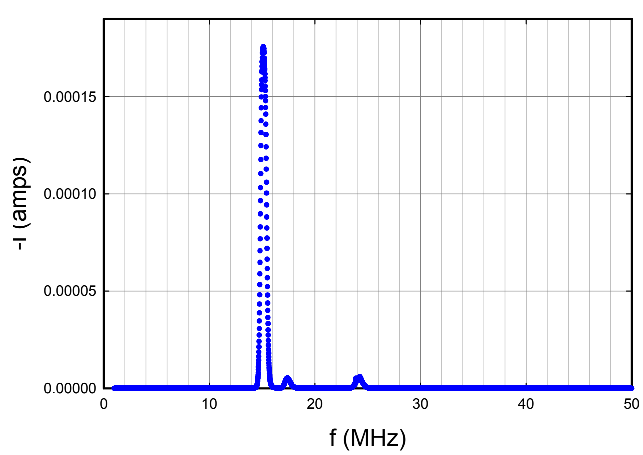

Figure 3. Example of a signal from the electrometer; I is the equivalent current and f is the drive frequency (the vibrational frequency is twice this drive frequency).

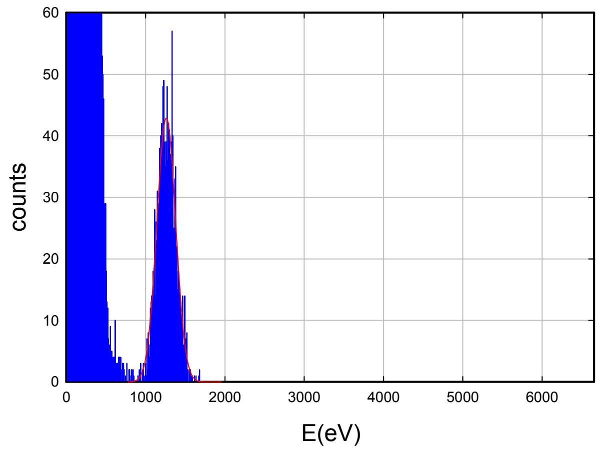

Figure 4. Example of a spectrum from the X-123 detector.

was not a result of electron emission from excited ^{201}Hg. In connection with the current signal we considered the possibility that up-converted vibrational energy was promoting electrons which could be picked up by air molecules and transported to the collector. In later tests it was found that there was no light evident near the foil surface in connection with the current signal, and also no obvious odor which would be expected if energetic electrons were ejected.

When the current signal was first seen, we were motivated to attempt measurements of X-rays. We saw features above 1 keV which appeared to be correlated with some of the expected vibrational resonances when Hg was on the surface; an example is shown in Fig. 4. It was possible to obtain clean spectrum with no surface Hg with no counts above 1 keV.

Perhaps it is understandable that results of this type could stimulate excitement. However, when we attended to writing up the experiment and results for publication, it became clear that there were unresolved issues associated with the calibration of the X-ray spectra. New calibration spectra were taken, and all of the data was systematically recalibrated and reviewed. In the process it became clear that the low energy feature near 1.5 keV that looks so promising in Fig. 4 would in some of the spectra show up at lower energies, and even at sufficiently low energy that it should have been absorbed by the 0.5 mil (12.5 μm thick) Be window (we would expect little X-ray transmission through this window below 1 keV). From this study it became clear that the detector was responding to something other than X-rays [24].

From discussions of this experiment with colleagues at MIT and elsewhere, it became clear that there was concern that the charge emission signals might also be artifacts. A similar experiment was set up at MIT and studied for some months, and in some cases signals showed up that were candidates for charge emission. With additional testing it was determined that these signals were noise. Near the end of this first MIT campaign we decided to develop a stringent test for charge emission at where the collector was connected through a capacitor to ground; in this case charge from the foil should charge up the capacitor and produce a measurable voltage. No significant voltage above a roughly 40 mV noise level was seen on a 5 pF capacitor at MIT.

Analogous experiments with a capacitor were carried out at SRI. In one set of experiments, a microfarad capacitor was used and monitored in real time with the electrometer running in voltage mode with a high impedance ($10^{14}\,\Omega$).

Strong signals were seen that appeared to provide a confirmation of the earlier current signals, showing a similar dependence on the drive frequency, and consistent with the sign and magnitude of the current signal. This appeared to provide confirmation of the charge emission result, and resulted in our submitting an abstract on our results for ICCF19.

A modified version of this test done subsequently led to different results, and has called into question the earlier current measurements. When a microfarad capacitor was again used for the test, but not monitored by the electrometer when the foil was driven, then no significant voltage was seen on the capacitor when measured after the excitation was turned off. Voltages up to about 10 V were seen with a 6 pF capacitor in this configuration, which would have been significant if real; but subsequent tests suggested that such signals were consistent with noise.

4. Discussion

It is clear from these studies that there is a learning curve associated with this kind of experiment. One issue is that using a linear amp to drive a low impedance reactive capacitor load ends up generating harmonics (at more than 10th order in experiments at MIT). The X-123 detector is very sensitive to noise; although at this point it has not been clarified whether the noise is due to RF, ultrasound, or due to something else. Measuring for charge emission is problematic in the presence of noise associated with the capacitive coupling excitation of the foil. The capacitor measurements suggest at this point that no charge emission has been seen; however it remains to be explained why monitoring the voltage on a microfarad capacitor with an electrometer that has an input impedance rated at 10^{14} ohms should lead to a false positive result.

Independent of how painful the learning curve associated with this new experiment might be, we are interested in how to implement a better version of the experiment generally, including reducing the noise, but also one which provides a better test. We are also concerned with the impact that a negative result so far on collimated X-ray emission has on the underlying theoretical picture.

It will be convenient to focus on the theoretical issue first. In the Introduction above we discussed briefly that a model had been developed over the years which describes both down-conversion relevant to the Fleischmann–Pons experiment, and up-conversion relevant to the Karabut experiment; it will be useful here to take this discussion a bit further. The early work on the models which focus on up-conversion and down-conversion required only that there be some coupling between the equivalent two-level systems standing in for the nuclear transitions, and the highly-excited oscillator which in the present discussion so far refers to the vibrational mode of the foil. When Karabut's experiment was analyzed making use of the relatively weak second-order coupling that would be expected between nuclear transitions involving phonon exchange, there was no way to obtain quantitative agreement between model and experiment. Subsequently, it became clear that agreement between theory and experiment could only come about if there were a much stronger coupling, which led to the adoption of a new fundamental Hamiltonian based on a relativistic description of the nuclei [26]. In this case there is a very strong first-order interaction between internal nuclear transitions and the center of mass momentum of the nuclei involved in the vibrations. Under normal conditions this first-order coupling is rotated out leaving a weak second-order relativistic correction that is usually ignored. However, we know from the analysis of the lossy spin-boson model (which is closely related algebraically) that the rotation becomes unhelpful for this kind of problem when the oscillator is strongly excited and when loss is present [27,28].

Once the new coupling became available, the theoretical up-conversion in Karabut's collimated X-ray experiments was revisited, multiple times. There were difficulties in the results from the modeling, which are worth considering here in light of the experimental results so far. As explained in the Introduction, the basic picture under consideration involved the up-conversion of vibrational quanta in the range of 50–250 MHz to produce nuclear excitation near 1.5 keV. In the normal up-conversion regime of the model relevant to this problem, the up-conversion power increases with a higher vibrational frequency, with stronger vibrations, with more nuclei involved, and with a strong low-energy nuclear transition. In the course of the campaign it became clear that the copper foil was not a particularly good candidate, since there are no strong low-energy nuclear transitions, and the number of nuclei present in a thin foil is limited. A response to this observation was to add the copper resonator to the SRI experiment in the hope of increasing the number of nuclei vibrating.

However, there remained headaches in the analysis. It became clear that even with the much stronger relativistic coupling, it was not possible to obtain agreement between theory and Karabut's experiment in the normal region of the model, assuming that a copper cathode was responsible for the up-conversion. Subsequently another pass was made through the analysis of the model which turned up an anomalous regime of the model, where the scaling laws were very different [29]. This anomalous regime of the model could be accessed when the coupling was extremely strong, or else if the nuclear transition energy is very low; and within this regime up-conversion is very much enhanced. Consequently, there seemed to be the possibility that a small cathode could provide the up-conversion needed to go from MHz to keV as long as it operated in the anomalous regime. For a copper cathode, since there are no relevant low energy nuclear transitions (the lowest energy excited state in ^{63}Cu is at 670 keV, and in ^{65}Cu is at 771 keV), this would imply that the very strong coupling required involved transitions with negative energy states. An analysis has not yet been done specific for negative energy state transitions, but we note that this is very much not an attractive model; we would only want to pursue this kind of approach if forced by experimental results that required it as the only possible option.

Given the unattractiveness of a model which relies on strong transitions to negative energy states for up-conversion in the anomalous regime of the model, it would be tempting to argue that a negative result with a copper foil rules out the scenario. Probably this would attribute too much weight to the present negative result. Nevertheless, a consideration of the different scenarios and issues suggests that we should re-think the model, the interpretation of the Karabut experiment, and we should also re-think the vibrating foil experiment. From this new perspective, our attention should be focused on stable isotopes with low-energy excited states, and we might expect these to be involved in the up-conversion process. Above we noted that ^{201}Hg and ^{181}Ta were candidates with the lowest energy transitions (at 1.56 and 6.24 keV); however, further down the list is ^{57}Fe with a transition energy of 14.4 keV [16]. This is significant due to the report of Kornilova et al. of the observation of collimated X-ray emission from a steel plate close to a water jet [30]. We recall that in Karabut's experiment the target chamber is made of steel.

This suggests that we should contemplate a revised interpretation for Karabut's collimated X-ray experiment. In this case, there are short (sub-ns) large amplitude (more than 50 kV) voltage spikes under normal operation in the high-current discharge which would be expected to couple to the cathode surface across the cathode fall region, and produce vibrations in the MHz regime that would vibrate a large number of ^{57}Fe nuclei in the roughly 80 pounds of steel target chamber. The vibrational frequency range of interest in this case is probably in the MHz region (note that the fundamental compressional mode in the 3 mm thick steel pate of Kornilova et al. is near 1 MHz). The conjecture is that vibrations in the large steel structure produces the up-conversion which is transferred to a small number [$O(10^{10})$] of impurity ^{201}Hg contaminant nuclei on the cathode surface, which then produces directional X-ray emission via a phased array emission effect.

In light of this new picture, a next generation version of the experiment should work with a larger sample that contains a suitable low-energy transition. Based on the interpretation of Karabut's collimated X-ray experiment, and also the interpretation of the water jet experiment of Kornilova et al., the focus should probably be on a steel sample; however also very much of interest would be a sample made of tantalum. We would like to drive this larger sample much harder, perhaps using a transducer matched to a resonance. To reduce noise we would like not to drive a reactive load with a linear amplifier. Given that the X-123 is so sensitive to noise, we would like to make use of X-ray film (keeping in mind that film has noise issues as well). We are continuing the experimental effort along these lines now at MIT.

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