Electron and positive ion acceleration with pyroelectric crystals

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The phenomenon of pyroelectric electron emission has been employed to develop miniature x-ray sources, such as the Cool-X by Amptek (www.amptek.com/coolx.html). The source strength of a pyroelectric x-ray generator is dependent on the emitted electron energy and current. Similarly, the source strength of a pyroelectric neutron generator will be dependent on the energy and production rate of deuterium ions in the fill gas. This paper summarizes our results in experiments directed toward creating high-energy electrons and positive ions with a pyroelectric source. Single-crystal sources are shown to produce positive ions with energies of up to 98 keV and electron energies of up to 143 keV. X-ray spectra are presented as proof that a paired-crystal source can increase electron energy to at least 215 keV. In addition, we offer independent verification of the "bunched" electron emission effect observed by [Brownridge *et al.*, Appl. Phys. Lett. **78**, 1158 (2001)]. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1884252]

I. INTRODUCTION

In 1974, Rosenblum *et al.* published a study of electron emission due to the heating of pyroelectric crystals.[1] Rosenblum *et al*. found that a $LiNbO_3$ crystal exhibited current densities of 10^{-10}–10^{-9} A/cm^2 when heated slowly from room temperature to 100 °C in a vacuum. They observed that the emission from the crystal was still significant when a 5-kV retarding potential was applied. In 1992, Brownridge published an article detailing the results from a pyroelectric x-ray generator.[2] He recognized that the energy of the electrons reported by researchers studying ferroelectric electron emission (FEE) was high enough to fluoresce a metallic target. He used the electrons emitted by pyroelectric $CsNO_3$ to fluoresce the *L*-shell electrons in gold, thereby beginning the study of x-ray generation via pyroelectric electron emission. Amptek, Inc., inspired by Brownridge's results, has since developed a battery-powered, pocket-sized, pyroelectric x-ray device with an x-ray endpoint energy of 36 keV, thus showing the commercial applicability of pyroelectric x-ray technology.[3]

The polarization of a crystal is the sum of the lattice cell dipole moments integrated over the volume of the crystal. A crystal is said to exhibit spontaneous polarization if this sum is nonzero without the application of an external electric field.[4] Pyroelectric crystals all show spontaneous polarization, although this polarization is usually masked by free charges in the air which accumulate on the crystal surface. The defining property of pyroelectric crystals is that their spontaneous polarization changes measurably when they are heated or cooled.[5] This is caused by the movement of atoms in the crystal lattice due to the change in temperature.[6] In the case of lithium tantalate ($LiTaO_3$) and lithium niobate ($LiNbO_3$), the pyroelectric crystals used in the experiments presented here, the lithium atom and tantalum (or niobium) atom in each unit cell move relative to the oxygen atoms which form the bulk of the lattice. This displacement relative to the oxygen atoms causes a change in the crystal polarization, hence the manifestation of the pyroelectric effect.

II. ELECTRON ACCELERATION

If a lithium tantalate or lithium niobate crystal is heated or cooled at atmospheric pressure, free charges in the air will readily compensate the change in polarization, thereby eliminating the creation of an electric field due to the change in polarization. If, however, the temperature change occurs in a vacuum, there are few free charges to mask the change in polarization, and we see the formation of a large electric field. Rosenblum *et al.*[1] reported an estimated electric-field strength of 1.35×10^7 V/cm for a lithium niobate crystal with $\Delta T = 75$ °C. Their estimate was a calculation based on the change in polarization as a function of the temperature change and the pyroelectric coefficient of $LiNbO_3$. (Our experimental results suggest that the electric field is roughly two orders of magnitude lower than this estimate.) This electric field is great enough to eject electrons from the dielectric layer at the surface of the crystal with a fraction of an eV of initial energy.[7] Once ejected, the electrons can be accelerated by the electric field to energies of up to 170 keV.[8] It is of note that the electric field which causes the electron emission will reverse when the temperature gradient on the crystal is reversed. Therefore, the $+z$ crystal surface, which emits electrons on heating, attracts electrons on cooling. The $-z$ crystal surface attracts electrons on heating and emits electrons on cooling.[9]

The change in polarization ΔP_s per unit area of a pyroelectric crystal is determined by the change in temperature ΔT times the crystal's pyroelectric coefficient γ,

$$\Delta P_s = \gamma \Delta T. \tag{1}$$

Therefore, for a crystal to be of practical use in a pyroelectric source, it must have a high pyroelectric coefficient, and a

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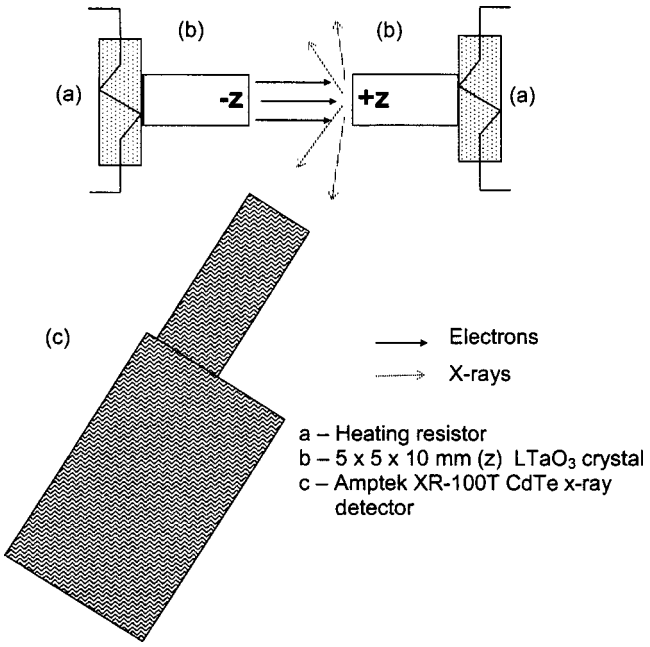

FIG. 1. Experimental geometry for bremsstrahlung energy measurements for a paired-crystal system. (A cooling phase is depicted.)

Curie temperature T_C which is high enough to allow the exposure of the crystal to a large ΔT.

Our experiments focused on the electron emission from $10(z)\times 5\times 5$ mm^3 $LiTaO_3$ pyroelectric crystals. In some experiments, two crystals were oriented to face each other in order to double the electron accelerating potential.

The electrons emitted by pyroelectric crystals can be detected and measured using surface-barrier detectors, such as the Canberra passivated implanted planar silicon (PIPS) A-300-19-AM, the detector used in our experiments. In circumstances where the use of a surface-barrier detector is not practical, such as the measurement of electron energy in a paired-crystal system, the energy of the (isotropically emitted) x rays from the electron interaction with high-Z materials may be used to estimate the electron energy. Figure 1 shows the setup for the detection of x rays from a paired-crystal system. It was common to place a steel absorber between the detector and the crystals to reduce dead time without significantly attenuating the high-energy photons.

For our measurements of electron energy from a single-crystal system, we placed the PIPS detector behind a 12 -mm-thick steel collimator with a 0.3-mm^2 aperture. A $10(z)\times 5\times 5$ mm^3 rectangular $LiTaO_3$ crystal was epoxied to two 39-Ω resistors connected in parallel, and was oriented such that the $-z$ surface of the crystal faced the aperture of the collimator. Figure 2 shows the experimental setup for the detection of electrons from a single pyroelectric crystal.

The electron energy from a pyroelectric electron emission starts in the 0–10-keV range, and then increases as the crystal cools to a maximum of $E\approx 143$ keV. Electrical discharge is a frequent occurrence and causes the electron energy and current to return to zero. If the discharge occurs in the middle of a cooling phase, the electron energy and current can return to levels similar to those before the discharge.

The highest electron energy we observed from a single

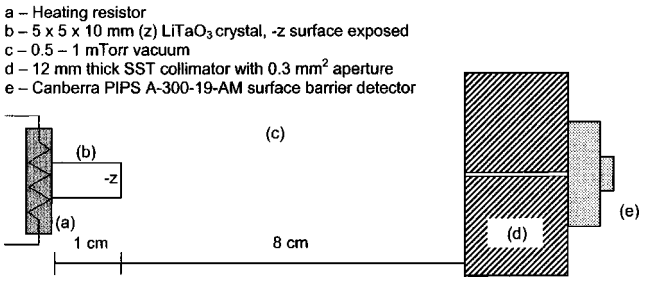

FIG. 2. Experimental geometry for the direct measurement of electron emission from a single-crystal pyroelectric source.

pyroelectric crystal using a surface-barrier detector was ~143 keV, as shown in Fig. 3. The full width at half maximum (FWHM) of the electron peak is ~8 keV. This includes broadening due to the change in electron energy due to crystal cooling, as well as the detector resolution, which was determined experimentally to be 3.8 keV for 88-keV photons. The highest x-ray energy observed from a paired-crystal system was ~215 keV, as shown in Fig. 4. The electron energy from the paired-crystal system must have been at least 215 keV to create bremsstrahlung of this energy.[10]

III. POSITIVE-ION PRODUCTION

An attractive potential use for pyroelectric crystals is in the production of neutrons via D–D fusion. For this potential to be realized, two steps must be overcome. First, it must be shown that the electrons emitted by the crystal ionize the fill gas, and that the ions are then accelerated by the crystal's electric field. Second, the energy of the positive ions must be greater than 30 keV, which is roughly the threshold energy for the D–D fusion reaction.

Since electrons carry a negative charge, the crystal's electric field will accelerate the electrons in a direction opposite to the acceleration of the positive ions. This means that when the positive ions are being accelerated toward the detector, the electrons will be accelerated toward the crystal.

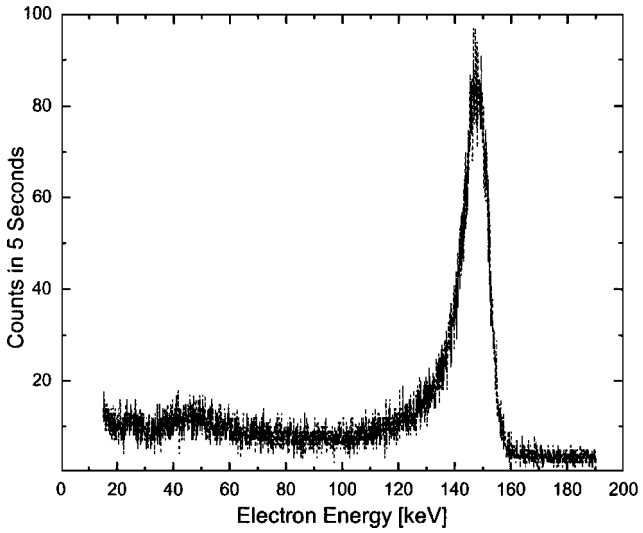

FIG. 3. Spectrum showing that the electrons emitted from a single-crystal pyroelectric crystal at a given time are nearly monoenergetic and can have an energy of ~143 keV.

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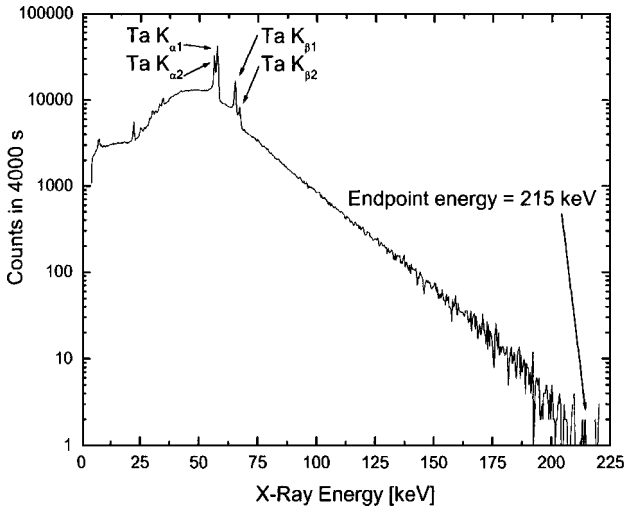

FIG. 4. X-ray spectrum showing the bremsstrahlung and characteristic x-ray production from a paired-crystal pyroelectric source. The ~215-keV endpoint energy shows that the maximum electron energy from a paired-crystal system must be at least 215 keV.

Since both types of pyroelectric crystals used in our experiments (niobium and tantalum) are reasonably efficient bremsstrahlung producers, this means that we will have to deal with a certain amount of noise from x rays whenever we want to measure a positive-ion spectrum. The x-ray noise can be reduced to a manageable level with the use of a pinhole collimator. This, of course, also reduces the amount of positive ions that we observe.

Brownridge and Shafroth claimed to have observed positive-ion production with a pyroelectric source.[11] Our attempt to verify the claim of Brownridge and Shafroth of positive-ion production consisted of the following experiment. A $5\times5\times10$ mm (z) $LiTaO_3$ crystal was epoxied to a resistor, with the $-z$ surface of the crystal being epoxied to the resistor and the $+z$ surface facing a surface-barrier detector. The detector was covered with a plastic collimator which was 6 mm thick and had a 1-mm-diameter aperture. This collimator was thick enough to reduce the count rate from a Cool-X x-ray source (which has an x-ray endpoint energy of 36 keV) by a factor of 33. Therefore, although the x rays generated in our system tend to have a higher energy than those from a Cool-X, the dead time of the detector could be reduced by attenuating the soft x rays from the crystals, which account for most of the x-ray spectrum. The distance between the crystal and detector varied, but was typically from 3 to 10 cm. Figure 5 shows the experimental setup for a positive-ion detection experiment. The crystal was heated for 10–30 min by passing a current through the resistor, reaching a maximum temperature of 140–170 °C. The crystal was then allowed to cool naturally, and "snapshot" spectra (spectra taken for a few seconds and then saved) were taken during the course of the cooling phase.

We detected a peak of about ~75–100 keV "riding" on the x-ray continuum. The energy of the peak changed as the crystal cooled, so the peak could not have been made by an x-ray fluorescence line in the crystal. Finally, a piece of electrical tape was used to cover the aperture in the collimator. When the tape was in place, the peak disappeared, but the

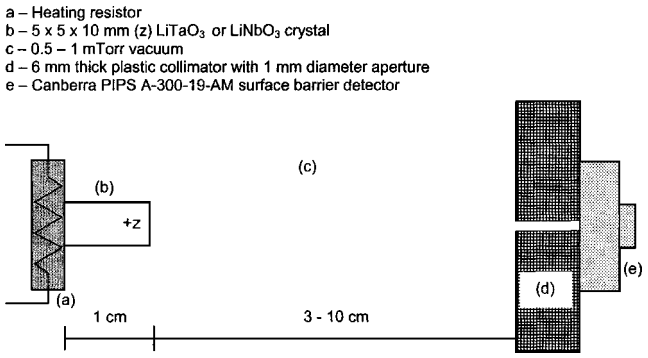

FIG. 5. Experimental geometry for the detection of positive ions accelerated by the potential from a pyroelectric crystal.

continuum remained. This experiment was repeated many times with and without the tape, and the same result occurred each time. Therefore, the continuum must have been from x rays, which can penetrate the electrical tape quite easily, while the high-energy peak must have been from positive ions, which can be blocked by a piece of tape. The peak could not have been from electrons; the crystal was polarized such that electrons were accelerated toward the crystal surface, as verified by the presence of the observed x-ray continuum in the positive-ion spectra. Figure 6 shows the positive-ion spectrum for our experiment with a $LiTaO_3$ crystal at two different temperatures during cooling.

This experiment was repeated with a 5-mm diameter $\times$10-mm-thick cylindrical $LiNbO_3$ crystal. The spectra taken during the cooling of the $LiNbO_3$ crystal show a low-energy peak at 18 keV, in addition to a high-energy peak which varies from ~30 to 60 keV. With the collimator hole covered with electrical tape, the 18-keV peak remains, while the high-energy peak disappears. Therefore, the 18-keV peak is from x-ray fluorescence of the niobium in the crystal (Nb has characteristic x-ray lines at 16.6 and 18.6 keV), while the high-energy peak is from positive ions. Figure 7 shows the positive-ion spectra from $LiNbO_3$ at two different temperatures during the cooling phase.

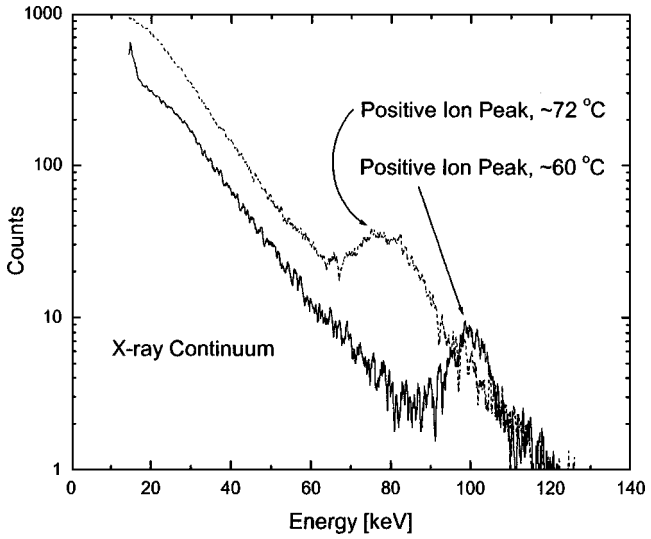

FIG. 6. Positive-ion spectra for a lithium tantalate driven source during cooling. The positive-ion peak changes in energy as the crystal cools.

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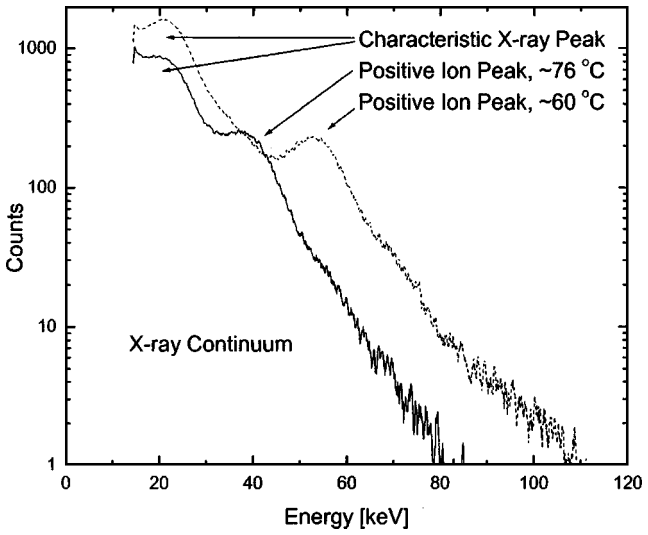

FIG. 7. Positive-ion spectra for a lithium niobate driven source during cooling. The peak which changes in energy is caused by positive ions, while the peak that stays stationary is due to characteristic x-ray emission from the electrons striking the crystal.

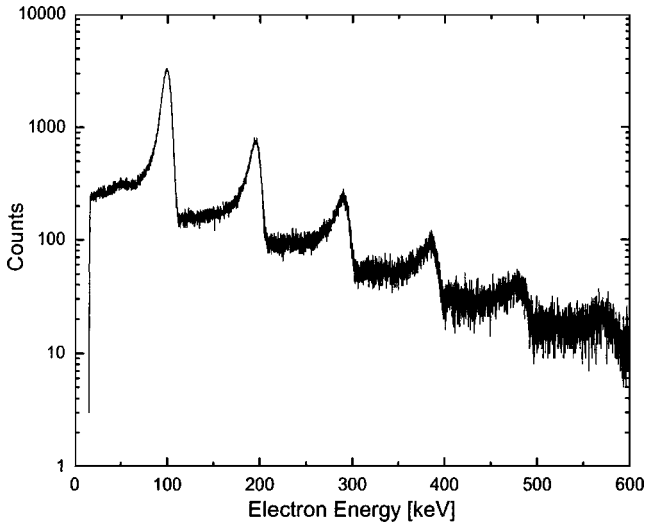

FIG. 8. Spectrum showing pileup peaks for electrons emitted by a $LiTaO_3$ crystal.

IV. REPEATED ELECTRON PEAKS

For one set of electron detection experiments, we used a 12-mm-thick steel collimator with a 1-mm^2 aperture instead of the 12-mm-thick steel collimator with a 0.3-mm^2 aperture. This allowed us to examine repeated peaks due to pulse pileup. Brownridge *et al.* reported repeated electron peaks from pyroelectric electron emissions that were of far greater size than would be expected due to pulse pileup[12] from random emission.

The exposure of a paralyzable electron detector to a high flux of monoenergetic electrons results in repeated peaks due to pulse pileup. The peaks will be separated by the true energy of the electrons. The size of the *r*th pileup peak is determined by the probability of *r* electrons interacting with the detector within its resolving time. For a random emission source in the paralyzable detector dead time model the true count rate *n* can be predicted from knowledge of the system dead time τ and the observed count rate *m*,

$$m = ne^{-n\tau}. \tag{2}$$

The probability of more than one particle interaction event being recorded by the detector as a single pulse is given by

$$P(x) = e^{-n\tau}(1 - e^{-n\tau})^x, \tag{3}$$

where $x-1$ is the number of particles recorded at the same time.[13]

Our detector was a Canberra PIPS A-300-19-AM. The resolving time of this detector and associated electronics was measured with an oscilloscope and found to be ~ 40 μs. We recorded a spectrum for 2 min, during a time in the cooling cycle where the electron energy and count rate were seen to be nearly constant. By comparing the number of counts in the pileup peaks observed in our electron measurements to that which is predicted by the paralyzable detector model, we find that the observed pileup greatly exceeds that which we would expect due to counting statistics alone. This result is in agreement with the observation of Brownridge *et al.* Figure 8 shows one of our spectra showing repeated electron peaks. Figure 9 is a comparison of our observed counts per peak for this experiment versus the counts per peak calculated with Eq. (3). The calculated and observed counts per peak were normalized at peak 1. This was done by using the counts in the first peak as the observed count rate *m* in Eq. (2) to calculate the actual count rate *n*. The actual count rate was then entered into Eq. (3) to calculate the expected counts per peak. At this time, it is unknown whether the large pileup peaks are due to "bunched" electron emission from the crystal itself or from the ionization of the fill gas and subsequent acceleration of the ionization electrons to the same energy as the ejected electrons.

V. CONCLUSIONS

The cooling of pyroelectric crystals in a vacuum of 10^{-4}–10^{-3} Torr can cause the formation of an electret state, accompanied by a potential of $>10^5$ V. This potential is sufficient to eject electrons from the crystal surface and accel-

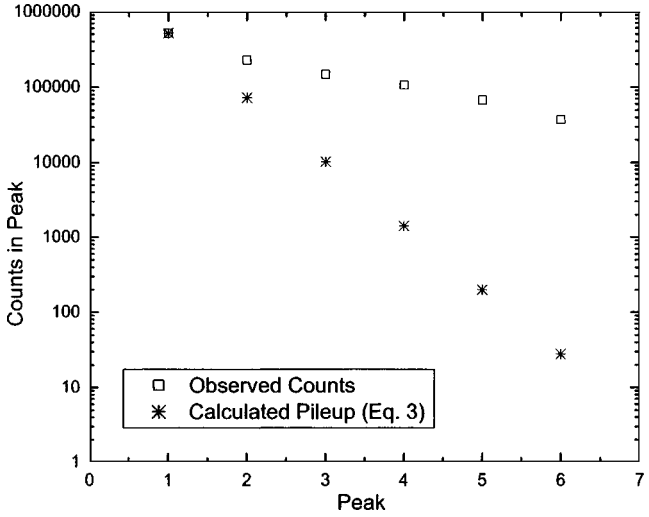

FIG. 9. Comparison of observed pileup peak area to calculated pileup expected from a random emission source.

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erate them to an energy of 143 keV for a single-crystal source, or at least 215 keV for a two-crystal source. Electron–ion pairs can also be created in the fill gas and accelerated by the electric field of the crystal. The electrons are accelerated in packets, which causes the observation of multiple "pileup" peaks, with the area of all peaks after the single electron peak being much greater than would be predicted due to the occurrence of pulse pileup from a random emission source.

The electrons emitted by the crystal can interact with atoms in the vacuum chamber fill gas to create electron–ion pairs. The positive ions are then accelerated in an opposite direction to that of the electrons, and can reach an energy of ~100 keV in a single-crystal source. It is unknown to what energy the positive ions in a two-crystal source are accelerated, but it can be assumed that the energy would be greater than 100 keV, due to our success in greatly increasing electron energy by using two-crystal electron sources.

If deuterons from a deuterium fill gas can be accelerated to energy similar to that observed with the ambient gas ions in our vacuum chamber, they can be accelerated into a deuterated target to cause D–D fusion, thereby creating a pyroelectric neutron source.

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