Electron Acceleration for X-ray Production Using Paired Pyroelectric Crystals

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Abstract - The exposure of pyroelectric crystals to a heating cycle in near-vacuum conditions has been shown to be a viable method of producing x-rays. The z-negative face of the crystal forms a positive charge as the crystal heats, and a negative charge as it cools. The opposite is true for the z-positive face. Literature has shown that this charge is great enough to accelerate electrons to energies of up to 170 keV. This phenomenon can be used to create small X-ray, electron and charged particle sources. X-rays are produced when the electrons in the ambient gas are accelerated by this field toward a target (when the exposed face of the crystal is negatively charged) or toward the crystal itself (when the exposed face is positively charged). In this experiment, z-cut lithium tantalate crystals with 5 mm x 5 mm surface area and thicknesses of 1, 2, 4, and 10 mm were placed in a vacuum and subjected to a thermal cycle. A 10 x10 x 0.5 mm crystal was also tested. The counts per thermal cycle and maximum energy of the x-ray spectra were shown to increase with crystal thickness. In addition, a system was explored in which two 10 mm crystals were arranged in series. This system was shown to double the maximum x-ray energy and yield are currently under investigation.

I. INTRODUCTION TO PYROELECTRIC X-RAY PRODUCTION

J. Brownridge et al. discovered that the exposure of a pyroelectric crystal in a vacuum to a heating cycle can be a useful phenomenon to create x-rays.¹ This technology has now been made available commercially by Amptek² in the form of a pocket-sized, battery powered x-ray device. We feel that this is an exciting new technology and that it is important to try to develop an understanding of the factors which influence the x-ray emission in a pyroelectric x-ray generator, and to suggest innovative methods to improve the x-ray yield and increase the endpoint energy.

Pyroelectric crystals are spontaneously polarized along an axis. In other words, they have a bulk dipole moment which exists in equilibrium conditions and does not sum to zero over the integrated volume of the crystal. This polarization is usually masked by the gradual accumulation of surface charges.³ The distinguishing trait of pyroelectric crystals is that the polarization of the crystal is altered dramatically with changes in the temperature of the crystal, such that the polarization is no longer masked and a large voltage can be observed across the crystal.⁴ The change in polarization per unit area with respect to the change in crystal temperature is known as the "pyroelectric coefficient," and is expressed in units of charge per unit area per degree Kelvin. Every pyroelectric crystal also has the property of transitioning into a non-polar state at a point known as the Curie temperature.³ In order for a crystal to be useful for x-ray production, its Curie temperature must be in a convenient range so the crystal can be subjected to a large temperature range without losing its polarization. The pyroelectric coefficient must also be great enough such that a sufficient potential for x-ray production can be achieved over some convenient temperature gradient.

The uncompensated polarization during the heating or cooling of the crystal causes the ejection of electrons from the dielectric layer on the surface of the crystal.⁵ Brownridge discovered that when a pyroelectric crystal is subjected to a thermal cycle in a vacuum, the electrons from the surface of the crystal and from the ambient gas can be accelerated by the potential of the crystal against a metallic target to create x-rays through x-ray fluorescence and bremsstrahlung effects.¹ Brownridge observed that when the -z surface of the crystal was exposed (where the z-axis is the axis of polarization) the characteristic x-rays of the crystal was cooled the characteristic x-rays of the target were observed.⁶

The electric potential of the crystal and target system, φ , may be found from Equation 1, where ΔP_s is the product of the change in temperature times the pyroelectric coefficient of the crystal, C_{cr} is the capacitance of the crystal, and C_{gap} is the capacitance of the gap between the crystal and the target.⁷

$$\varphi = \frac{\Delta P_s}{C_{cr} + C_{gap}} \tag{1}$$

We can draw two immediate conclusions from this relationship. Since the potential is inversely proportional to the capacitance, and the capacitance is inversely proportional to the thickness of the medium, we should expect that any increase in the thickness of the crystal will result in an increase in the potential of the system. We should also expect this increase to be linear. Equation 1 suggests that the same effect can be obtained by moving the target further away from the crystal. This is true, but the change in potential due to target distance was found to be negligible in comparison the change due to the change in crystal thickness as long as the target was more than ~ 1 mm away from the crystal. This corresponds to the large difference in capacitance between a lithium tantalate crystal and a vacuum of the same thickness.

Lithium tantalate (LiTaO₃) is a useful pyroelectric crystal to use in x-ray production experiments due to its high pyroelectric coefficient (~190 μ C * m⁻² * K⁻¹)³ and high Curie temperature (665°C)⁸. Pyroelectric x-ray generators employing lithium tantalate crystals also can operate in an easily obtainable pressure range, which was reported by Shafroth et al. to be within the operating range of mechanical fore pumps.⁹ This statement was confirmed experimentally for this paper.

The objective of this research was to test the dependence of x-ray yield and maximum x-ray energy on crystal thickness, to qualitatively discuss the effect of ambient gas pressure and the heating rate on x-ray production, and to test the effect of a second crystal on the maximum x-ray energy and intensity.

II. EXPERIMENTAL SETUP

Our experiments were conducted in a vacuum chamber connected to a mechanical pump capable of achieving pressures of ~20 mTorr. A valve was located between the pump and the chamber allowing the pressure of the chamber to be regulated within ranges of ± 2 mTorr. An Amptek CZT-100T detector was located within the chamber. A K-type thermocouple was connected to an HP 3457A multimeter which was read by a GPIB card and provided temperature input to a LabView temperature control program. The program then provided output in the form of a signal from an HP 3324A function generator coupled to a purpose-built voltage-to-current converter. This supplied current to the thermoelectric heater / cooler [THC] which provided the heating cycle to the crystal.

The THC was attached to a copper and aluminum heat sink, and the bottom edge of the THC rested on a plastic insulating base. Crystals were attached to the THC with thermally conductive, electrically isolating grease. The z+ face of the crystal faced the detector window, so that we would see target emission on heating. А grounding strip was placed between the THC and the crystal. The detector window was approximately two centimeters away from the THC surface. A 25 µm thick copper target was located between the crystal and the detector window, and was very close (~1 mm) to the detector window. See Figure 1 for a diagram of the experimental setup. For the tests of the effect of crystal thickness on x-ray production, crystals with thicknesses of 0.5 mm, 1 mm, 2 mm, 4 mm, and 10 mm were tested. The 0.5 mm crystals were polished and were 10 mm on the x and y dimensions, while the rest of the crystals were not polished and were 5 mm on the x and y dimensions.



Figure 1 - Chamber Setup for Thickness Tests, side view

The LabView program received a user input temperature profile, and raised or lowered the current to the thermoelectric cooler based on whether the crystal needed to be heated or cooled to match the command profile. In this way we were able to consistently reproduce the same thermal cycle, and were able to accurately compare the yield from different thermal cycles. After every thermal cycle the chamber was vented and allowed to sit for a few minutes. This was done because we experienced a reduction in electron emission over consecutive thermal cycles, an observation that was also made by Rosenman.⁴

The experimental setup was modified for the oneversus two-crystal tests by the addition of a U-shaped copper bracket, as shown in Figure 2. The center of the bracket was attached to the grounding strip and thermoelectric heater, and a 5 x 5 x 10 mm crystal was attached with grease to each "wing" of the bracket. The crystals were arranged such that the +z face of one crystal was facing the -z face of the other crystal. A thin wafer of circuit board material was used as a spacer so the crystals would not stick together due to electrical attraction. The crystals were held up by insulating brackets at the center of each crystal instead of with a plastic base. The spacing between the crystals was 10 mm for the two-crystal system. To test the yield from the one-crystal system, a wafer board shim was inserted between the crystal and the bracket so that the crystal would remain electrically and thermally isolated, but would still be in position to act as a target for the emission from the other crystal. This reduced the distance between the crystals to ~8 mm for the one-crystal tests. The detector window was ~5 cm away from the front of the crystals. No metallic target was used in the two crystal the bremsstrahlung and fluorescence x-rays system:

observed by the detector in experiments with that system were from interaction of the electrons with the crystals themselves.



Figure 2 - Two Crystal System, Top View

III. EFFECT OF CRYSTAL THICKNESS ON X-RAY YIELD

Lithium tantalate crystals with thicknesses of 0.5 mm, 1 mm, 2 mm, 4 mm, and 10 mm were tested to determine the effect of crystal thickness on x-ray energy and yield. We created several heating cycles in the LabView program, each of which went on a linear path from 293°K to 373°K and back to 293°K over some period of time. Each crystal thickness was tested at each heating rate at some set of consistent experimental conditions (i.e., constant pressure range, distance from detector, target distance). Based on these tests, the optimum heating rate was determined. We then kept the heating profile constant for each thickness and tested the crystals at different pressure ranges.

III.A. Effect of Heating Rate

The heating rate had some effect on the x-ray production from a crystal at a given pressure range. The only severe effects occurred when the crystal was heated very rapidly or very slowly, which resulted in dramatic reduction of the counts observed per thermal cycle. The drop in intensity at rapid heating rates may be due to the poor thermal conductivity of the crystal, which means that the surface facing the detector may not have experienced the same thermal cycle as the surface contacting the THC. It may also have dropped from once crystal face grounding to the other through the plastic base or the gas due to a rapid formation of potential across the crystal. During very slow thermal cycles, the crystal may have had time to re-mask its polarization through the accumulation of free charges from the gas, and by gradual electrical conduction through the crystal or the plastic base. We observed that the optimum heating rate in terms of total counts per thermal cycle was usually about half the rate at which the crystal could no longer react to the entire thermal cycle.

III.B. Effect of Chamber Pressure

After testing for the effect of the heating rate on x-ray production, we tested the effect of the pressure inside the chamber. Past research performed by Brownridge suggests that the electron emission from a pyroelectric crystal in a dilute gas is subject to a gas multiplication effect.¹⁰ He directed the electron emission from a pyroelectric (lithium niobate) crystal toward a zinc sulfide screen and observed the spot made by the electrons interacting with the screen. As pressure increased, the beam spot became wider and brighter, until the pressure reached 8 mTorr, at which point the intensity dropped to zero. This suggests that the gas pressure has a great effect on x-ray intensity.

Our tests of pressure effect on intensity were performed by comparing the gross counts over a given thermal cycle at different pressures for each crystal thickness. We found that the optimum pressure increased as the crystal thickness decreased, with the exception of the 0.5 mm thick crystal. This crystal was polished and was larger in surface area than the other crystals, and so may not have been completely comparable. Our observed optimum pressure ranges are shown in Table I.

Table I - Optimum Pressure vs. Thickness

Crystal Thickness	Optimum Pressure
[mm]	[mTorr]
0.5	51 ± 3
1	73 ± 5
2	53 ± 3
4	46 ± 6
10	36 ± 5

III.C. Effect of Crystal Thickness

The capacitance of a layer is given by Equation 2, where C is the capacitance, ε is the dielectric constant of the medium, ε_0 is the permittivity of free space, A is the area of the layer, and d is the thickness of the layer.

$$C = \varepsilon \cdot \varepsilon_o \cdot A/d \tag{2}$$

Therefore, since the dielectric constant of lithium tantalate is 51, we should expect the gap capacitance term in Equation 1 to be negligible compared to the crystal capacitance term. For this reason, we expected the energy of emission to increase as a linear function of crystal thickness alone, as can be seen by substituting Equation 1 into Equation 2 to obtain Equation 3:



Each crystal thickness was tested at its optimum pressure and heating rate to determine the dependence of x-ray pressure and yield as a function of thickness. Figure 3 shows a spectrum obtained by subjecting a 4 mm thick crystal to one heating cycle and is included as an example of typical x-ray emission from a transmission geometry pyroelectric x-ray generator. Notice the depression in the spectrum caused by the 8.98 keV absorption edge in copper and the peaks caused by XRF at 8.05 and 8.90 keV.



Figure 3 - Emission from 5 x 5 x 4 mm Crystal, One Heating / Cooling Thermal Cycle

Several typical spectra were selected for each thickness for the purpose of comparison. The endpoint energy of the spectra as a function of crystal thickness is shown in Figure 4. Since the observed endpoint energies of the 0.5 mm and 1 mm crystals are very close to the 8.98 keV absorption edge in copper, it is possible that the endpoint energies of the 0.5 mm and 1 mm samples were misrepresented as being lower than they actually are. One can see, however, that the maximum x-ray energy depends on the thickness of the crystal in a nearly linear relationship.



Figure 4 - Endpoint Energy vs. Crystal Thickness

The x-ray yield per thermal cycle also increased as a function of crystal thickness, as shown in Figure 4.



Figure 5 - Counts per Cycle vs. Crystal Thickness

IV. ONE - VERSUS TWO-CRYSTAL SYSTEM

We tested two crystals in series to try to accelerate the electrons with the combined electric field of two crystals in an attempt to double the maximum x-ray energy. The experimental setup for the two-crystal system was described earlier. The spectra shown in Figure 6 were taken at 32 to 39 mTorr with a 0.27° K / s heating cycle that went from 20°C to 100°C, stayed at 100°C for 100 seconds, and then returned to 20°C. Only the heating phase of the thermal cycles is shown. The addition of the second crystal roughly doubled the endpoint energy of the system, raising it from 21 keV to 45 keV.



Figure 6 - One- versus Two-Crystal System

As Figure 6 shows, the x-ray yield from two thermal cycles in a one-crystal system is roughly equivalent to the x-ray yield from one cycle in a two-crystal system. Therefore, in addition to the increase in x-ray endpoint energy, the two-crystal system has twice the intensity as a similar one-crystal system.

V. CONCLUSIONS

These experiments show that the maximum energy and x-ray yield from a pyroelectric x-ray generator are dependent on the thickness of the crystal, and that the relationship between the thickness and the maximum energy and the x-ray yield is nearly linear. The maximum energy can be doubled with the addition of a second crystal.

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