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Applications of pyroelectric particle accelerators

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Abstract

The discovery of pyroelectric X-ray generation in 1992 by Brownridge has led to a recent surge of interest in the use of the pyroelectric effect as a means of producing useful radiation. By heating or cooling a pyroelectric crystal such as lithium tantalate (LiTaO₃) in a vacuum, a potential on the order of 100 kV can be generated. This potential is great enough to eject electrons from the crystal for the production of characteristic or bremsstrahlung X-rays, or to cause field ionization near a tip mounted to the crystal. By using the combined fields of two polarized crystals, the acceleration potential can be doubled, with one crystal acting as a particle emitter and the other crystal serving as a target. Such a paired-crystal system was used to generate X-rays with energies of greater than 200 keV, and can be used to fluoresce the K shell of thorium (Z = 92). An alternative use of pyroelectric sources is the field ionization of a dilute gas. If the positively-charged crystal is used to ionize a deuterium gas, and the target crystal is coated with deuterated target, the deuterium ions can be accelerated into the target at high enough energy to cause D–D fusion. Results verifying the production of D–D fusion neutrons from a pyroelectric source will be presented. Future applications of pyroelectric accelerator technology, such as the use of the electron beam for materials testing, will also be discussed. © 2007 Elsevier B.V. All rights reserved.

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1. Introduction

Pyroelectric crystals have been shown to be useful materials for the production of low-cost, portable X-ray sources [1,2]. Recently, their use has been extended to the production of neutrons via D–D fusion [3,4]. This report shows that the surface charge on the crystals can be predicted with a simple differential equation, and that this charge can be used to produce an external electron beam. We also demonstrate fusion neutron production using pyroelectric crystals, and suggest methods of improving the neutron yield.

2. Electron emission

Due to the pyroelectric effect, temperature changes cause charge to be developed on the faces of a pyroelectric

crystal perpendicular to the axis of polarization. The surface charge density σ , neglecting losses, is given by [5]:

$$\sigma = \gamma \cdot \Delta T,\tag{1}$$

where γ is the material-specific pyroelectric coefficient and ΔT is the change in temperature. Consider the z– surface of a LiTaO₃ crystal, which becomes positively charged during heating and negatively charged during cooling. During a cooling phase, charge is lost from the surface due to: relaxation current through the crystal; screening charges from the surrounding environment, which mask the accumulated charge; electron emission from the negatively-charged crystal surface; and electron emission from surrounding materials incident on the positively-charged surface. Since the crystals are poor conductors, the relaxation current is a very weak effect, with a time constant on the order of 10⁴ s. The effect of screening charges can be minimized by heating and cooling the crystal in a vacuum. Therefore, we are left with one important phenomenon, that of electron emission from

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the negatively-charged surface of the crystal. Finally, we obtain a simple differential equation for the surface charge:

$$\frac{\mathrm{d}q}{\mathrm{d}t} = \gamma \cdot A \cdot \frac{\mathrm{d}T}{\mathrm{d}t} - i_{\mathrm{FE}},\tag{2}$$

where i_{FE} is the field emission current and A is the crystal area.

In a simple experiment, we mounted a 5 mm (dia.) \times 10 mm (z) LiTaO₃ crystal such that it emitted electrons toward a 25 µm Be window during cooling. The window was 2 cm from the emitting surface of the crystal. A copper Faraday cup was used to measure the current from the crystal, which was recorded in LabView. An Amptek XR-100CT CdTe X-ray detector was used to record the X-ray emission from the Faraday cup. The extrapolated endpoint energy from these spectra was used to estimate the electron energy striking the target.

We next sought to compare our experimentally-measured current to the mathematical model laid out in Eq. (2). Field electron emission from a conductor is governed by the Fowler–Nordheim equation [6]:

$$i_{\rm FE} = 6.2 \times 10^{-6} \frac{(\mu/\phi)^{1/2}}{\mu + \phi} \cdot F^2 \exp\left[-6.8 \times 10^7 \frac{\phi^{3/2}}{F}\right], \qquad (3)$$

where *F* is the electric field strength in V/cm, μ is the Fermi energy in eV, and ϕ is the work function in eV. The physics of electron emission from a polarized dielectric such as a pyroelectric crystal differs somewhat from the tunneling of a conduction electron through a triangular potential barrier considered in the Fowler–Nordheim model. However, we can keep the basic form of the model and apply it to our differential charge equation to obtain:

$$\frac{\mathrm{d}q}{\mathrm{d}t} = \gamma \cdot A \cdot \frac{\mathrm{d}T}{\mathrm{d}t} - a \cdot F^2 \exp\left[-\frac{b}{F}\right],\tag{4}$$

where *a* and *b* are fitting parameters. The pyroelectric contribution to the charge balance equation, $\gamma \cdot A \cdot \frac{dT}{dt}$, was found by using the slope of the temperature profile measured in LabView as the $\frac{dT}{dt}$ term and multiplying by the area of the crystal and the known pyroelectric coefficient of LiTaO₃, $\gamma = 190 \ \mu C/(m^2 \text{ K})$ [7].

Fig. 1 shows a plot of the crystal current and temperature versus time. The temperature was measured at the base of the crystal, so there is a delay between the beginning of the cooling phase (as measured by the thermocouple) and the current emission. In addition to the measured current, corrected for attenuation through the Be window, we show the current predicted by Eq. (4), with fitting parameters $a = 2.5 \times 10^{-19}$ and $b = 7.05 \times 10^{-6}$. These fitting parameters are shown to be a good approximation to the measured current.

An important feature of this experiment was that the electron current was measured outside of the vacuum chamber. In previous experiments with pyroelectric crystals, the target had always been inside the vacuum chamber. Fig. 2 shows the transmission probability for electron beams of



Fig. 1. Calculated field emission current versus current measured with a Faraday cup outside a thin Be window.



Fig. 2. Probability of electron transmission through a 25 μ m Be window, as calculated by MCNP. Error was < 0.25% at each data point used to plot this curve.

different energy incident on a 25 μ m Be window, as calculated in MCNP. As Fig. 1 shows, electrons were able to penetrate the window steadily for nearly half an hour. X-ray measurements showed that the maximum electron energy was approximately 120 keV. This technique provides for the use of a sealed pyroelectric electron source for materials testing and treatment, or possible medical uses. For example, a 0.25 cm diameter LiTaO₃, during a single cooling phase, can deliver approximately 500 Gy of radiation to a 130 μ m deep volume of skin (assuming 100 keV electrons, diverging to a 0.5 cm diameter beam).

3. Neutron production

In 2005, Naranjo et al. demonstrated that fusion neutrons could be produced using pyroelectric crystals [3]. They did this by cryogenically cooling a LiTaO₃ crystal, and then heating it to ionize deuterium gas with a metallic tip mounted on the crystal. Deuterons produced near the tip were accelerated away from the crystal with a ~100 kV acceleration potential into an erbium deuteride target, causing fusion. We were able to confirm the possibility of producing neutrons with a pyroelectric source using a paired-crystal source. In this configuration, two 10 mm thick \times 20 mm diameter LiTaO₃ crystals were separated by 2.5 cm, with the z+ surface of one crystal facing the z- surface of the other. It was shown through X-ray measurements that a similar source could accelerate electrons to >200 keV [8].

The crystal with the exposed z+ surface was covered with a copper disc attached with electrically conductive epoxy. A 70 nm radius, 3 mm long tungsten catwhisker tip was soldered to the disc. The other crystal was covered with deuterated polystyrene, $-(C_8D_8)_{n-}$ (later the target material was changed to deuterated polyethylene, $-(C_2D_4)_{n-}$). In this configuration, ions would be produced near the 70 nm tip and accelerated into the deuterated plastic target as the crystals were cooled from their maximum temperature of ~ 130 °C to room temperature. The neutron detector was a 5 in. \times 3 in. Eljen Technology EJ-301 liquid scintillator 8.1 cm from the center of the target, and at angle perpendicular to the target plane. Pulse shape discrimination was used to eliminate photon counts.

It was expected that the advantage gained in acceleration potential over the experiment used by Naranjo et al. would result in an increased neutron yield. However, we found that our yield was roughly an order of magnitude weaker than that observed at UCLA [4]. Part of this discrepancy is due to the difference in crystal size: we used a 20 mm dia. crystal instead of a 30 mm dia. crystal, and therefore reduced the charge available for ion formation by 2.25. However, this still does not account for decreased neutron yield.

One way in which we can alter the original experiment is to change the conductivity of the epoxy attaching the copper disc and metal tip to the pyroelectric crystal. We have always used electrically conductive epoxy, which was intended to allow the free flow of charge to the tip to ionize the deuterium gas. However, as shown in Fig. 3, we can also ionize deuterium gas with a non-conductive epoxy attaching the tip and disc to the crystal, since the electric field of the crystal is still able to induce a field at the tip. Fig. 3 is a direct measurement of deuterium ions using a silicon detector (Canberra A-300-19-AM PIPS) [8] inside the vacuum chamber. We expect the use of non-conductive epoxy as the interface between the tip and the disc to reduce the ionization efficiency, but thereby increase the system potential, causing an increased cross section for fusion.

Another possible way of increasing the neutron yield is to use a slightly broader ionizing tip. If we model the tip as a small sphere at a potential of 100 kV, and assume that deuterium will be ionized in an electric field which provides at least the ionization potential of deuterium (15.4 V) over



Fig. 3. Deuterons generated by a 70 nm tip mounted with a nonconductive epoxy to a LiTaO₃ crystal. Collimature aperture size = 1/ 50 mm², distance to crystal = 7.5 cm. Deuterons were measured with a Canberra A-300-19-AM PIPS detector mounted in the vacuum chamber.



Fig. 4. Critical volume for deuterium ionization in the vicinity of a charged sphere. Volume of charged sphere excluded.

the approximate size of a deuterium atom (10^{-10} m) , we can estimate that all atoms within a radius where the field remains above a critical value of $F_{\text{crit}} = \frac{15.4 \text{ V}}{10^{-10} \text{ m}} = 1.54 \times 10^{11} \text{ V/m}.$

The maximum electric field increases as the tip size decreases. However, the larger the tip, the slower the electric field decreases as a function of radius. A critical tip size value exists where the field is high enough to ionize the gas, but the field strength decreases slowly as a function of radius to allow a large ionization volume. Therefore, our model predicts that the optimum tip size may be much larger than the 70 nm catwhisker tips we had previously used in our experiments (see Fig. 4). While it is obvious that the assumption that our tips behave like charged spheres is a gross simplification, this result stresses the importance of optimizing the tip size in pyroelectric fusion experiments.

4. Summary

Pyroelectric crystals can be used to create many types of portable radiation sources. By heating or cooling paired LiTaO₃ crystals in vacuum, one can generate X-rays of over 200 keV or generate D–D fusion neutrons with a low-voltage power supply. A single pyroelectric crystal can create a beam of ~100 keV electrons which can be extracted from a vacuum chamber through a thin Be window to test materials in open air. Finally, we have shown that a simplified version of the Fowler–Nordheim equation can be used to mathematically predict the current produced by such an electron source.

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