

High-energy x-ray production with pyroelectric crystals

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(Received 20 July 2004; accepted 28 March 2005; published online 11 May 2005)

The invention of pyroelectric x-ray generator technology has enabled researchers to develop ultraportable, low-power x-ray sources for use in imaging, materials analysis, and other applications. For many applications, the usefulness of an x-ray source is determined by its yield and endpoint energy. In x-ray fluorescence, for example, high-energy sources enable the excitation of the *K*-shell x-ray peaks for high-*Z* materials as well as the lower-energy *L*-shell peaks, allowing more positive sample identification. This report shows how a paired-crystal pyroelectric source can be used to approximately double the endpoint x-ray energy, in addition to doubling the x-ray yield, versus a single-crystal source. As an example of the advantage of a paired-crystal system, we present a spectrum showing the fluorescence of the *K* shell of thorium using a pyroelectric source, as well as a spectrum showing the fluorescence of the *K* shell of lead. Also shown is an x-ray spectrum with an endpoint energy of 215 keV. © 2005 American Institute of Physics. [DOI: 10.1063/1.1915536]

I. INTRODUCTION

Pyroelectric crystals, when heated or cooled, exhibit a change in the polarization proportional to the crystal's pyroelectric coefficient times the magnitude of the temperature change.¹ In atmospheric conditions, the abundance of free charges causes a masking charge to rapidly form on the polarized surfaces, preventing the change in the polarization from reaching levels useful for x-ray production. In a vacuum, however, the result of heating a pyroelectric crystal can be the ejection of electrons from the crystal surface² and their acceleration to energies of nearly 200 keV. If these energetic electrons strike a metallic target, they are sufficiently energetic to create bremsstrahlung and fluorescence x rays. This x-ray generation technique was discovered by Brownridge.³

Pyroelectric crystals exhibit spontaneous polarization under equilibrium conditions. This polarization is usually masked by a screening charge. However, when a pyroelectric crystal is heated or cooled in the absence of a readily available screening charge (i.e., in a vacuum), the spontaneous polarization ΔP_s can be changed by an amount proportional to the change in temperature ΔT times the pyroelectric coefficient of the material,¹ γ :

$$\Delta P_s = \gamma \Delta T. \quad (1)$$

The change in the polarization due to the heating of a pyroelectric crystal competes with the relaxation current through the crystal and the loss of charge due to electron emission.

The relaxation time constant for a pyroelectric crystal dictates the rate at which some amount of charge at the crystal surface will diffuse through the crystal to counteract the buildup of charge due to the pyroelectric effect. The relaxation time constant τ is given by¹

$$\tau = \frac{\epsilon \cdot \epsilon_0}{\sigma}, \quad (2)$$

where ϵ is the relative permittivity of the crystal, ϵ_0 is the permittivity of free space, and σ is the bulk conductivity of the crystal. The loss of surface charge Q through relaxation is given by the differential equation:

$$\frac{\partial Q}{\partial t} = -\frac{1}{\tau} Q. \quad (3)$$

Since the bulk conductivity for lithium tantalate and lithium niobate is very low, the relaxation time constant is much longer than the typical heating cycle for a pyroelectric x-ray generator. (For example, for lithium niobate, $\tau=7720$ s.¹ The relaxation time for lithium tantalate is assumed to be similar.) Therefore, charge relaxation through the crystal bulk is an inefficient way of neutralizing the change in the polarization due to the pyroelectric effect.

The most efficient means of neutralizing the surface charges caused by a temperature change is the emission of electrons from the crystal. Rosenblum *et al.*² measured the emission current from a LiNbO₃ crystal heated from room temperature to 100 °C to be 10^{-10} – 10^{-9} A/cm². The potential acting on the emitted electrons can be found by modeling the crystal and x-ray target as a two-capacitor system (see Fig. 1). If the crystal-target system is modeled as two capacitors in parallel, the system potential is given by¹

$$\varphi = \frac{\Delta P_s}{C_{cr} + C_{gap}}, \quad (4)$$

where φ is the system potential, C_{cr} is the capacitance of the crystal, C_{gap} is the capacitance of the gap between the crystal and the target, and ΔP_s is the change in the spontaneous polarization due to the pyroelectric effect [see Eq. (1)]. The capacitance of the crystal is defined as

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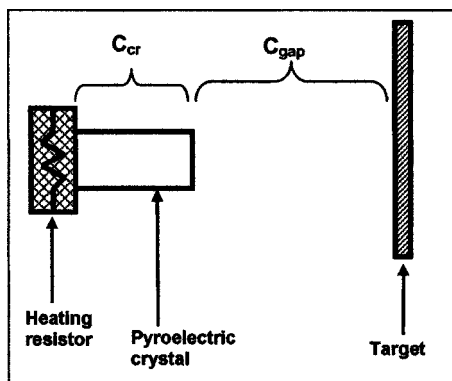


FIG. 1. Representation of a pyroelectric crystal and its metallic bremsstrahlung target as two parallel capacitors.

$$C_{\text{cr}} = \epsilon_{\text{cr}} \cdot \epsilon_0 \cdot \frac{A_{\text{cr}}}{d_{\text{cr}}}, \quad (5)$$

where A is the area of the crystal, d is the thickness of the crystal, ϵ is the relative permittivity of the crystal, and ϵ_0 is the permittivity of free space. The capacitance of the vacuum layer between the crystal and the target can be found in a similar manner. By substituting Eq. (5) into Eq. (4), we obtain

$$\varphi = \frac{\Delta P_s}{\epsilon_{\text{cr}} \cdot \epsilon_0 \cdot \frac{A_{\text{cr}}}{d_{\text{cr}}} + \epsilon_{\text{gap}} \cdot \epsilon_0 \cdot \frac{A_{\text{gap}}}{d_{\text{gap}}}}. \quad (6)$$

It is apparent that an increase in the thickness of either the crystal or the gap will increase the potential of the crystal-target system, and therefore should increase the energy of the electrons incident on the target. Since the relative permittivity of lithium tantalate and lithium niobate is much greater than the relative permittivity of a vacuum, the effect of an increase in the vacuum layer thickness is negligible compared to the effect of a similar increase in the thickness of the crystal. As an example, the relative permittivity of lithium tantalate along the z -axis is 46. Therefore, the capacitance of a 1-cm-thick LiTaO_3 crystal is 46 times greater than the capacitance of 1 cm of vacuum, and we should expect the energy of the emitted electrons to increase as a roughly linear function of the crystal thickness.⁴ It is worth mentioning that we have had little success in increasing the x-ray energy from a LiTaO_3 -driven pyroelectric source by increasing the crystal thickness past 10 mm, due to a saturation of the energy versus crystal thickness curve which we do not yet understand.

The invention of pyroelectric x-ray generation technology by Brownridge has made possible the development of ultraportable x-ray sources which are ideal for classroom and field use, such as the Amptek Cool-X.⁵ These portable x-ray devices are especially useful for analyzing materials through x-ray fluorescence (XRF). Pyroelectric sources are advantageous in portable XRF devices because they emit harmful radiation only when turned on. XRF is caused by the excitation of an atomic electron from its ground state to an excited state, followed by the deexcitation of the electron accompanied by the emission of a characteristic x ray. For this reason, in order to observe a characteristic x ray of a particular en-

ergy, we must have a source photon of at least that energy in order to excite the electron. The amount of energy required to excite a particular group of characteristic x rays increases as a function of atomic number.

Our research at Rensselaer Polytechnic Institute (RPI) has led to a very effective approach to increasing the maximum x-ray energy from a pyroelectric source.⁴ Instead of using the electric field produced by the temperature change of a single pyroelectric crystal to accelerate electrons into a metallic target, we orient two identical lithium tantalate (LiTaO_3) pyroelectric crystals with opposite polarity to point at each other, and simultaneously expose them to the same temperature cycle. In this way, we are able to double the magnitude of the electric field acting on the source electrons, and therefore approximately double the x-ray endpoint energy created when the electrons from the source crystal collide with the tantalum atoms in the target crystal.

Our method of using paired pyroelectric crystals to create high-energy x rays has enabled us to fluoresce metals at higher energies than we were able to accomplish using single-crystal sources. Of particular significance is our ability to use a two-crystal source to fluoresce the K -shell lines of lead ($Z=82$) and thorium ($Z=90$).

II. X-RAY PRODUCTION USING PAIRED PYROELECTRIC CRYSTALS

Two lithium tantalate crystals, measuring $10(z) \times 5 \times 5 \text{ mm}^3$, were epoxied to 20- Ω heating resistors. One crystal was epoxied with its $+z$ surface attached to the resistor and its $-z$ surface exposed, the other was epoxied with its $-z$ surface epoxied to the crystal and its $+z$ surface exposed. The resistors were wired to a terminal strip mounted to a rigid piece of perforated board. The resistor wires were bent to allow orientation of the crystals.

Power to the heating resistors was provided by a dc power supply, usually set to 6 V. Typically, the crystals were heated for 10 min and then cooled for 10–15 min. The maximum temperature for each experiment was between 172 and 176 °C. For measurements with a single-crystal system, the wire providing power to the heating resistor to the crystal with the exposed $+z$ surface was disconnected. In this way, we were able to faithfully reproduce the geometry of the paired-crystal system, since the vacuum chamber did not have to be opened, nor the crystals moved, to reconfigure the system to perform a single-crystal system test. No metallic target was used to produce x-rays in this experiment. During cooling, when our spectra were collected, the crystal with the exposed $+z$ surface acted as a target for the electrons emitted by the crystal with the exposed $-z$ surface. We collected our spectra with an Amptek CR-100T-CdTe x-ray detector. Figure 2 is a schematic depiction of the geometry for the paired-crystal versus single-crystal experiments.

Figure 3 shows the result of using paired crystals instead of a single crystal. In the paired-crystal system, the endpoint energy of the x-ray spectrum is approximately 184 keV, versus only 107 keV for the single-crystal system. The K -shell characteristic x-ray peaks for tantalum are also more clearly visible in the paired-crystal system, which is further evidence

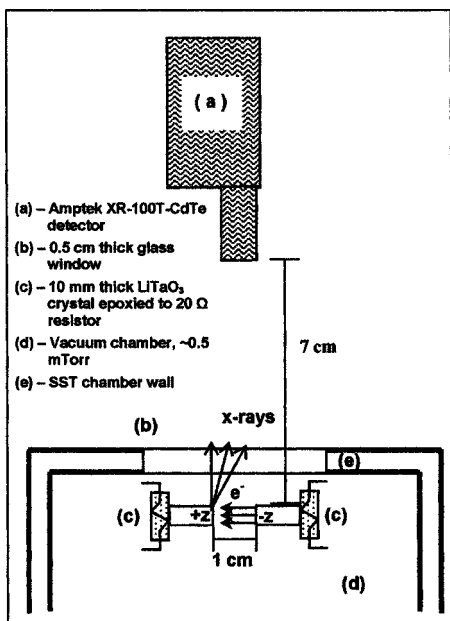


FIG. 2. The experimental geometry for single- vs paired-crystal system comparison.

of its effect on high-energy x-ray yield. In addition to the increase in energy due to the use of a second crystal, the total x-ray yield is approximately doubled, from an average of about 261 counts/s during a cooling cycle for a single crystal system to an average of 518 counts/s during a cooling cycle for a paired crystal system. (The count rate varied greatly in each experiment, and the low average count rates reported here are due to the spectrum recording time, which was set for 800 s regardless of how quickly the count rate dropped to near zero.)

It has been our experience that the grounding of the rear (i.e., nonemitting) surface of the crystals can cause a slight increase in the observed x-ray endpoint energy. In order to determine the highest achievable energy with a paired-crystal x-ray generator, we utilized the experiment geometry shown in Fig. 4. The pyroelectric x-ray generator consisted of two grounded $10(z) \times 5 \times 5 \text{ mm}^3$ rectangular LiTaO_3 crystals,

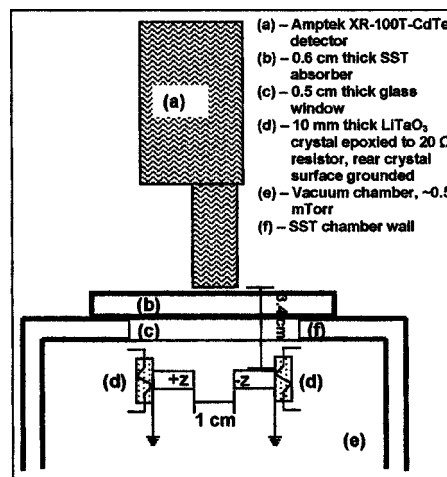


FIG. 4. The experimental geometry used to determine the maximum x-ray energy from a paired-crystal pyroelectric x-ray generator.

separated by 1 cm. A 0.6-cm steel absorber was placed between the vacuum chamber window and the CdTe x-ray detector. This was done to absorb most of the low-energy x rays without causing significant attenuation of the high-energy x rays. The absorber allowed us to operate with the detector very close to the window to avoid geometric attenuation of the high-energy x rays without causing pulse pileup due to an excessive maximum count rate. The maximum count rate observed in these experiments was ~ 1400 counts/s with an average count rate of 415 counts/s. As in previously described experiments, the average count rate was taken over a recording time of 800 s, regardless of how quickly the emission from the source dropped to near zero.

Figure 5 shows the sum of five cooling phases in this geometry, during which the crystals were cooled from approximately 165°C to room temperature. The maximum x-ray energy is shown to be 215 keV.

III. FLUORESCENCE OF HEAVY METALS

Brownridge published results showing the fluorescence of the *K*-shell lines of gold ($Z=79$),⁶ and later was able to

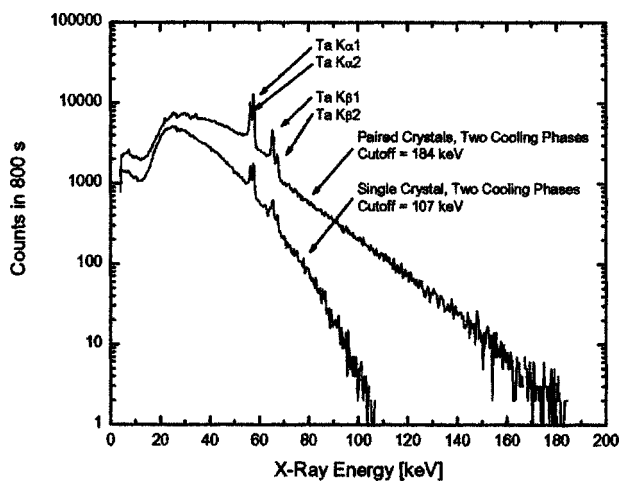


FIG. 3. Two spectra showing the increase in x-ray energy by using paired pyroelectric crystals.

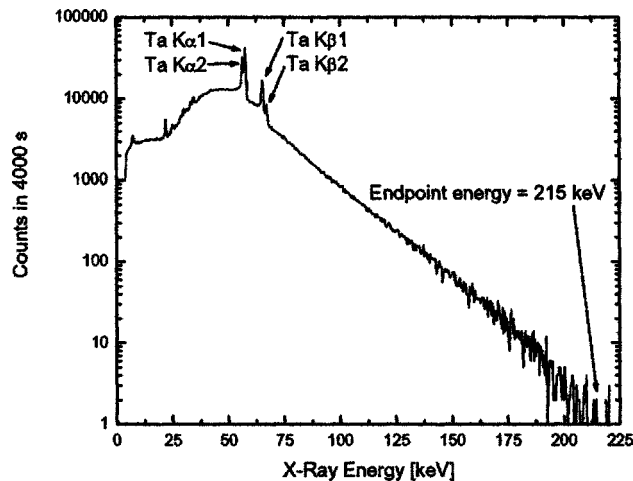


FIG. 5. The x-ray spectrum showing the highest observed x-ray energy from a pyroelectric source, 215 keV.

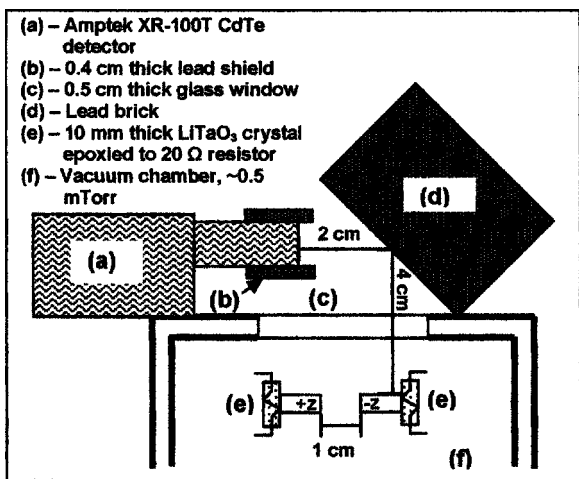


FIG. 6. The experimental geometry for the fluorescence of a lead brick with x rays produced by a paired-crystal pyroelectric x-ray generator.

fluoresce lead ($Z=82$), although those results are not yet published.⁷ The positive results pertaining to the increase in the maximum x-ray energy by using paired crystals led us to attempt to reproduce Brownridge's results, and then to fluoresce materials with higher atomic number than lead.

Figure 6 shows our geometry for lead fluorescence. The x rays produced via bremsstrahlung and XRF in the target crystal penetrate the 0.5-cm glass window, and some of them hit the lead brick. The glass window on the vacuum chamber is thick enough to stop all of the source electrons, and therefore no bremsstrahlung is produced in the brick, only characteristic x rays. 0.4 cm of lead shields our detector from the bremsstrahlung produced by the target crystal, so that the only photons observed in the spectrum are due to the fluorescence of the lead brick.

Figure 7 shows the spectrum obtained from summing the spectra from the cooling phases from two of our lead fluorescence experiments. In addition to the L -shell characteristic x rays, the spectrum clearly shows the K_{α} (72.8 and 74.9 keV) and K_{β} (84.9 and 87.2 keV) characteristic x-ray peaks.

After successfully fluorescing lead, we attempted to

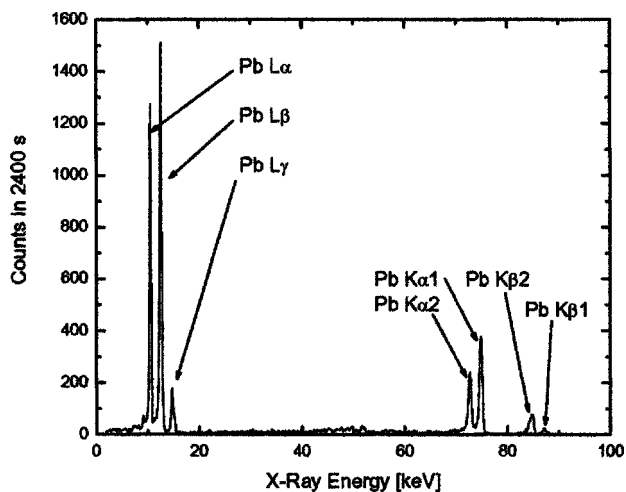


FIG. 7. The x-ray spectrum showing the fluorescence of lead using x rays generated by a paired-crystal source.

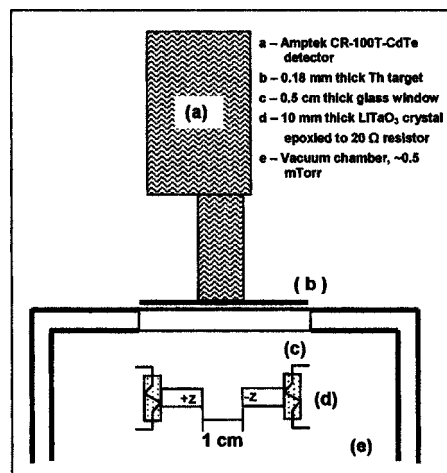


FIG. 8. The experimental transmission geometry for the fluorescence of a thin thorium target with x rays produced by a paired-crystal pyroelectric x-ray generator.

fluoresce thorium ($Z=90$). Our thorium sample was approximately 0.2 mm thick, and could therefore be penetrated easily by ~ 100 keV x rays. Instead of using a reflection geometry similar to that of in our lead fluorescence experiments, we took advantage of the thinness of the thorium sample by using a transmission geometry, shown in Fig. 8, in which x rays from the target crystal would pass through the glass window and strike the thorium target. Some x rays would then fluoresce the thorium, and some of these thorium characteristic x rays would be able to penetrate the sample and be measured by the detector.

A challenge presented by the thorium fluorescence experiment is the natural radioactivity of the sample, which in our case was ^{232}Th . To compensate for the x ray and gamma radiation emitted by the thorium target, we recorded a 4000-s background spectrum immediately prior to recording the 800-s (or 1000-s) spectra in which the pyroelectric x-ray system was used to fluoresce the target. A paired-crystal system

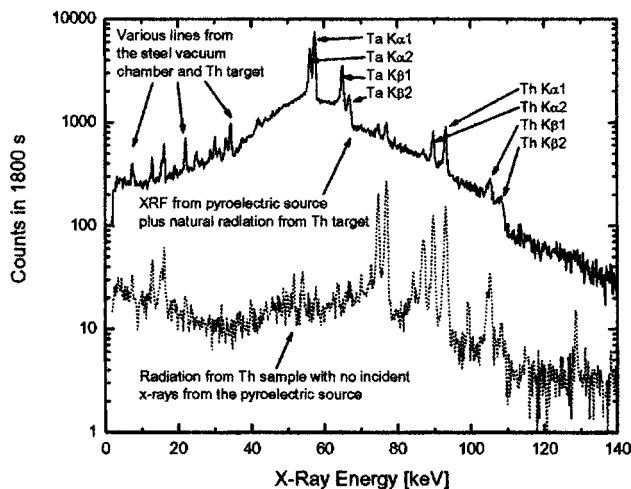


FIG. 9. Comparison of the background x ray and gamma ray spectrum due to the natural radioactivity of the thorium sample and a spectrum showing both the background radiation and the radiation due to the x rays produced by the cooling of a paired-crystal pyroelectric x-ray source. These spectra were both normalized to 1800 s.

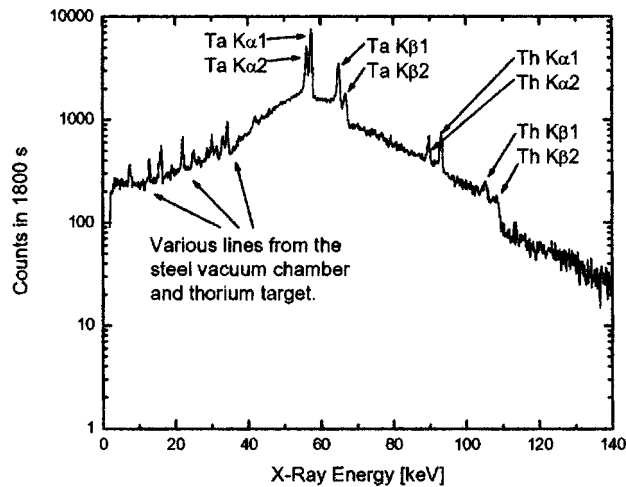


FIG. 10. The spectrum showing the result of subtracting the background spectrum from the spectrum taken during the cooling of the pyroelectric source.

using two 10-mm-thick LiTaO_3 crystals was heated to 172°C and then cooled. The x-ray spectrum transmitted through the thorium foil was recorded during the cooling phase. Two such spectra were added together, and the thorium background spectrum (normalized to 1800 s, the total recording time for the two fluorescence x-ray spectra) was subtracted to give a spectrum representing the net x-ray yield due to the x rays from the pyroelectric x-ray generator. Figure 9 shows the thorium fluorescence and background spectra for this experiment. Figure 9 shows the thorium fluorescence and background spectra for this experiment. Figure 10 shows the net x-ray spectrum after the subtraction of the background radiation from the thorium sample. The spectrum in Fig. 10 clearly shows the K_α (90.0 and 93.4 keV) and K_β (105.6 and 108.5 keV) x rays from fluorescence with the paired-crystal pyroelectric source. The tantalum characteris-

tic x-ray lines and bremsstrahlung continuum are also visible because the thorium target was not thick enough to efficiently attenuate them.

IV. CONCLUSIONS

The results shown in this paper are evidence of the advantage to be gained by generating x rays with a paired-crystal pyroelectric x-ray generator instead of a single-crystal system. The use of a second crystal with inverse orientation to the emitting crystal results in nearly doubling the potential acting on the emitted electrons. Evidence for this effect is given by the x-ray spectrum in Fig. 3, in which we show that the x-ray energy was increased from 107 to 184 keV. This remarkable increase in energy gives us sufficient high-energy x-ray yield to fluoresce the K -shell lines in elements with higher atomic number than before, including thorium, which is the highest- Z material for which K -shell fluorescence has yet been achieved with a pyroelectric source. In addition to the increase in energy from using paired crystals instead of a single-crystal system, the x-ray yield is also approximately doubled. Finally, the 215-keV endpoint energy of the x-ray spectrum in Fig. 5 is the highest x-ray energy published for a pyroelectric x-ray generator to this date. Such a source is very attractive for portable XRF systems, since the source can be turned off after use, requires little shielding, and can be powered by a dry cell battery.

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⁵www.amptek.com/coolx.html

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