Key Engineering Materials Vol. 582 (2014) pp 174-177
Online available since 2013/Sep/10 at www.scientific.net

doi:10.4028/www.scientific.net/KEM.582.174

Improvement of the Stability of X-ray Emission by Thermal Excitation of a Pyroelectric Single Crystal

Fumihiko Naruse[1,a], Hiroyuki Honda[1,a], Yoshikazu Nakanishi[1,a], Shinji Fukao[1,a], Yuuki Sato[1,b], Yoshiaki Ito[2,c] and Shinzo Yoshikado[1,a]

[1]Graduate School of Science and Engineering, Doshisha University, Kyotanabe 610-0321, Japan
[2]Institute for Chemical Research, Kyoto University, Gokasho, Uji 611-0011, Japan

[a]syoshika@mail.doshisha.ac.jp, [b]yusato@mail.doshisha.ac.jp, [c]yosi@elec.kuicr.kyoto-u.ac.jp

Keywords: X-rays, $LiTaO_3$, pyroelectricity, thermal excitation, creeping discharge, stability of X-ray intensity, periodicity of heat cycle

Abstract. X-ray emission using pyroelectric crystals is intermittent, and has low intensity and stability. One of the factors for low stability is related to creeping discharge, due to the accumulation of surface electric charges that change in response to the temperature. The time dependence of the net amount of electric charge was investigated by changing the cycle period of the crystal temperature. The stability of the X-ray emission is demonstrated to be strongly dependent on the temperature cycle period.

Introduction

Spontaneous polarization of a pyroelectric crystal such as $LiNbO_3$ or $LiTaO_3$ is dependent on the temperature of the crystal, and electric charge appears on the crystal surface by heating or cooling of the crystal. This pyroelectricity causes intense electric fields to appear around the crystal when the temperature changes. The electric fields accelerate electrons toward the crystal or a target facing toward the crystal, and the chamber walls. X-rays are then emitted by the impact of electrons against these surfaces [1,2].

At the present time, conventional X-ray tubes are generally used as an X-ray source. An X-ray tube requires an electron source, a high voltage power supply, and a cooling system for the target. This configuration restricts miniaturization and reduces the X-ray emission efficiency. In contrast, a method of X-ray emission that exploits the thermal excitation of a pyroelectric crystal requires only a change of the crystal temperature in a vacuum; therefore, both miniaturization and an improvement of the X-ray emission efficiency can be achieved. However, this method also has disadvantages, such as unstable X-ray intensity compared to that obtained with an X-ray tube. These problems make the realization of practical applications difficult. Therefore, if these disadvantages can be addressed, then it is expected that this method could be applied to radiation therapy or portable diagnostic equipment.

One of the factors that causes unstable X-ray intensity is related to electrostatic creeping discharge along the surface of a crystal (hereafter, referred to as discharge), due to the accumulation of electric charges on the crystal surface. Therefore the stable emission of X-rays for a long time requires the crystal temperature to be periodically changed to form intense electric fields. The net amount of electric charge on the surface is determined by both the polarization charges and adsorbed charges supplied from ionized ambient gas. When the net amount of charge exceeds a certain threshold, discharge from one crystal surface to another crystal surface occurs and the charges are neutralized and disappear. Accordingly, sudden cessation of the X-ray emission occurs periodically. To avoid such discharge and achieve the stable emission of X-rays, the initial amount of electric charge on a crystal for each temperature cycle must be equal.

It is suggested that the stability of the X-ray emission is dependent on the cycle period of the crystal temperature, because the frequency of the discharge is varied with respect to the cycle period. In this study, the time dependence of the net amount of electric charge was simulated using a simple model [3]. Furthermore, the stability of the X-ray emission was investigated by changing the period of the temperature cycle. In addition, the amount of charge accumulated on the crystal was estimated.

Experimental

Figure 1 shows a schematic diagram of an experimental apparatus used to emit X-rays. A nonstoichiometric $LiTaO_3$ single crystal with dimensions of 10×10×5 mm^3 was used. The electrical surfaces (*z* surfaces), which were perpendicular to the *c*-axis, were polished to a mirror finish. The chamber was made from stainless steel (SUS 304). An oxygen-free copper foil with a thickness of 20 μm was used as the target and was placed 15 mm above the negatively charged surface (–*z* surface) of the crystal. The positively charged surface (+*z* surface) of the crystal was attached to a Peltier device using conductive paste (Fujikura Kasei D-550). The chamber, target and lower surface of the crystal (+*z* surface) were electrically grounded. The crystal temperature was controlled by the application of triangular voltages to the Peltier device with a duty cycle of 50%. The temperature range ΔT, was fixed at approximately 40 °C. The temperature change period *L*, was varied from 200 to 2000 s. The ambient gas was air and the pressure in the chamber was approximately 10^{-4} Pa. The temperature of the +*z* surface was measured using a thermistor sensor. X-ray emission was measured using a Si-PIN X-ray detector (Amptek XR-100CR) through a 0.3 mm diameter pinhole formed in a 0.5 mm thick lead sheet. The distance between the target and the detector was 8 mm. The effects of the detector efficiency and the pinhole were corrected. All measurements were performed continuously over approximately 50 periods.

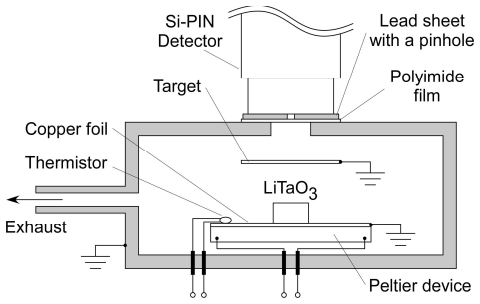


Fig.1. Schematic diagram of the experimental apparatus.

Results and Discussion

Figure 2 shows the energy spectra of X-rays per period for L = 1000 s. Cu K_α and Cu K_β X-rays originated from the target, and Ta L_α X-rays from the crystal. The X-rays emitted during the cooling phase were more intense than those emitted during the heating phase. Figure 3 shows the average count rate of X-ray photons for various values of *L*. These measurements were performed in the order of (d), (b), (a), (c), (f), and (e). *L* of 200, 250, and 300 s was measured over 1000 periods each. A periodic change of the X-ray intensity was observed. In the period during which the average count rate changed significantly, X-ray emission suddenly stopped. This is caused by the discharge, because when discharge occurs, a pulse-like current from the target, light emission around the crystal, and branch-like traces on the surface of the crystal have been reported [4]. Therefore, it is suggested that the discharge occurs periodically. The frequency of the change varied according to *L* and the intensity of X-rays was stable when *L* was 300 or 1000 s. Furthermore, both the intensity and the stability of X-rays were similar between (b) and (c), where *L* was 300 s, even with the insertion of the unstable emission (a) with *L* of 200 s.

Figure 4 shows a part of the cyclic characteristics of the average count rate of X-ray photons per period during the cooling process for *L* of 250 s, where the horizontal axis represents the total time. All discharges occurred during the cooling process. After the discharge, the average count rate increased linearly up to approximately 10 cycles and then asymptotically reached the value observed before discharge. The X-ray intensity is determined by the number of electrons that collide with the target. One model that may explain such a transition of the count rate is proposed. Firstly, when the average count rate increases linearly, it is thought that the transition of the electric charge distribution on the –*z* surface of a crystal becomes that shown in Fig. 5(a); when the crystal is heated, its spontaneous polarization decreases with the increase in temperature. However, positive charges adsorbed on the –*z* surface cannot follow the change of polarization, and are therefore desorbed. Thus, the –*z*

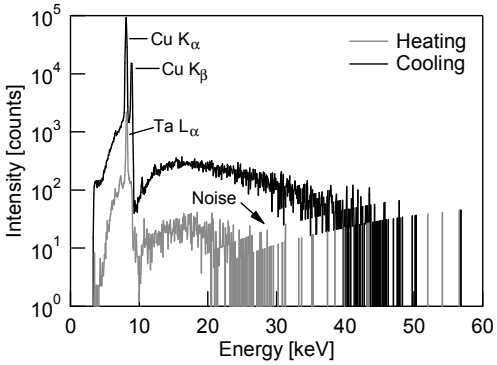
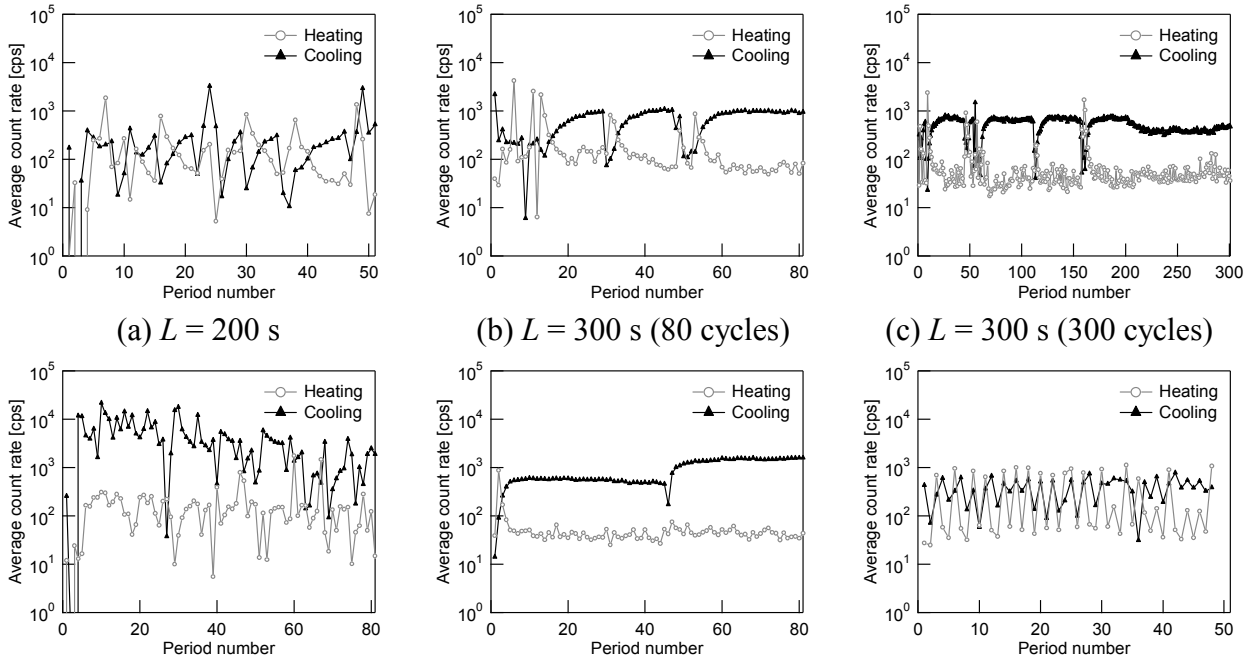


Fig.2. Energy spectra of X-rays per period for L = 1000 s.



(a) L = 200 s (b) L = 300 s (80 cycles) (c) L = 300 s (300 cycles)

(d) L = 600 s (e) L = 1000 s (f) L = 2000 s

Fig.3. Average count rate of X-ray photons for L = (a) 200, (b) 300 (80 cycles), (c) 300 (300 cycles), (d) 600, (e) 1000, and (f) 2000 s.

surface is positively charged, which induces electron adsorption. During the subsequent cooling phase, some of these electrons are accelerated toward the target and the others remain adsorbed on the $-z$ surface. It has been reported that the accumulation of positive charges occurs on the z surface [4]. The amount of positive charges increases as the temperature cycle is repeated, which results in an increase of both the electrons that collide with the target and those that are adsorbed on the surface. However, when the amount of the positive charges exceeds a certain threshold, it suddenly becomes difficult for electrons to desorb from the surface. It is speculated that this is due to strong attractive forces acting on the electrons by the electric field caused by the electric dipoles formed by adsorbed positive charges and electrons. Thus, the net amount of negative charge on the $-z$ surface becomes large during the cooling process by the restriction of electron desorption. When the intensity of the electric field due to these negative charges exceeds a certain threshold, discharge occurs on the surface and X-ray emission suddenly stops, as shown in Fig. 5(b).

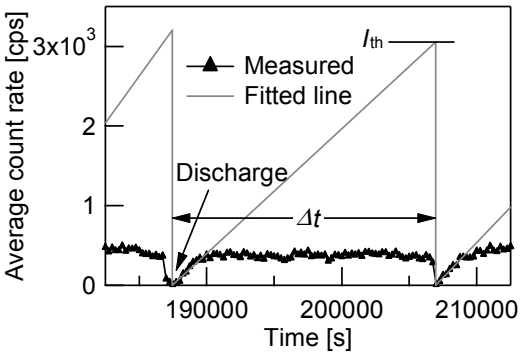


Fig.4. Average count rate of X-ray photons during the cooling process for L = 250 s.

Estimation of the threshold surface charge density was performed. It is speculated that the average count rate during the cooling process is proportional to the number of electrons desorbed from the surface during the period when the count rate increases linearly, as shown in Fig. 4. A straight line fitted to the average count rate after discharge was extrapolated to the time when the next discharge occurred. Here, the discharge interval Δt, and the extrapolated average count rate I_{th}, are defined as shown in Fig. 4. In addition, a variable X [cps] was introduced as a quantity that corresponds to the total amount of electrons supplied to the $-z$ surface. Furthermore, it is speculated that the surface charge density threshold σ_{th}, is given by:

$$\sigma_{\mathrm{th}} = \frac{1}{2} C(X - I_{\mathrm{th}})\Delta t \qquad (1)$$

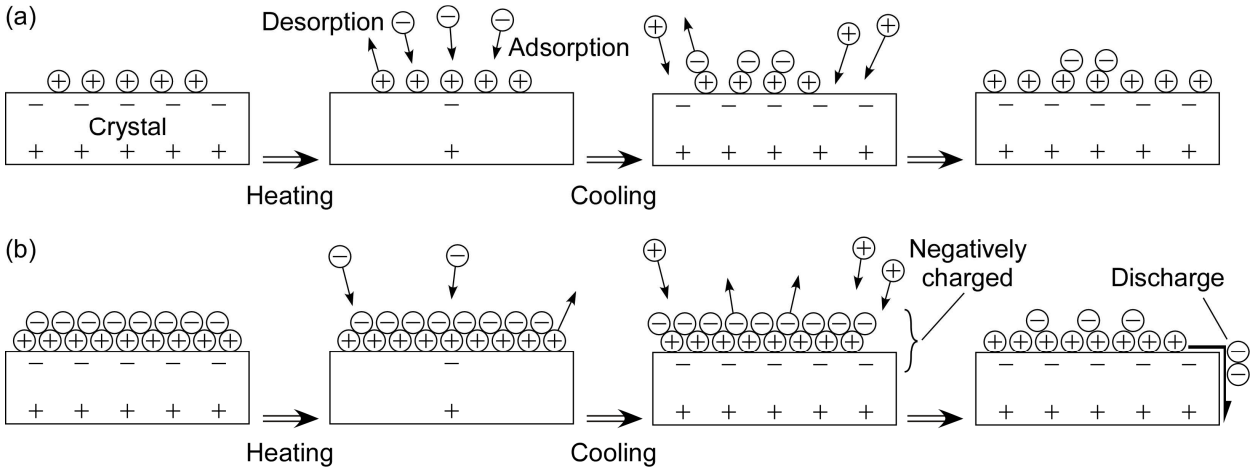


Fig. 5. Schematic diagram of the mechanism for charge accumulation on the crystal surface; (a) after discharge, and (b) just before discharge.

Table 1. Number of discharges and average values for Δt and I_{th}.

L [s]	Number of discharges	Δt [s]	I_{th} [cps]
200	15	1.23×10^4	1.1×10^3
250	9	2.22×10^4	4.0×10^3
300	16	1.67×10^4	2.1×10^3

Table 2. Solutions for Eq. 1.

Combination of equations	X [cps]	σ_{th}/C [counts]
(i)	7.8×10^3	4.1×10^7
(ii)	1.1×10^4	8.3×10^7
(iii)	4.4×10^3	2.1×10^7

where C is a constant. Table 1 shows the number of discharges and the average values of Δt and I_{th} for L of 200, 250, and 300 s. When L was set as 250 s, the number of discharges was the minimum and Δt was the maximum. Using these values, the three simultaneous equations of Eq. 1 for (i) L = 200 and 250 s, (ii) 250 and 300 s, and (iii) 300 and 200 s were solved by making X and σ_{th}/C unknowns. Table 2 shows the solved values, where the X and σ_{th}/C values for each combination of equation were almost the same magnitude.

Summary

When the period of temperature change L is varied, the long-term stability of the X-ray intensity is significantly changed. X-rays could be emitted over 20,000 s without discharge by selecting an appropriate value of L. Furthermore, the amount of accumulated charges on the crystal surface was estimated.

References

[1] J. D. Brownridge: Nature Vol. 358 (1992), p. 287

[2] J. D. Brownridge and Sol Raboy: J. Appl. Phys. Vol. 86 (1999), p. 640

[3] T. Z. Fullem and Y. Danon: J. Appl. Phys. Vol. 106 (2009), p. 074101

[4] H. Honda, Y. Nakanishi, S. Fukao, Y. Sato, Y. Ito and S. Yoshikado: Key Eng. Mater. (2012), in press.

Electroceramics in Japan XVI
10.4028/www.scientific.net/KEM.582

Improvement of the Stability of X-Ray Emission by Thermal Excitation of a Pyroelectric Single Crystal
10.4028/www.scientific.net/KEM.582.174