Generation of ionizing radiation from lithium niobate crystals

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Abstract. The work done experimentally explores generation of electron and x-ray radiation in the process of heating and cooling monolithic and iron-doped crystals of lithium niobate. Iron doping to the concentrations in the range of $10^{23}$ m$^3$ was carried out by adding ferric oxide into the melt during the process of crystal growth. The research into radiation generation was performed at 1-10 Pa. The speed of heating from -10 to $10^7$ °C was 10-20 degrees a minute. Current pulses appeared at 17, 38, 56, 94, 98, 100, 105, 106, $10^7$ °C with the interval of 1-3 minutes. The obtained electron current increased in direct proportion to the crystal surface area. The maximum current was 3mA at the design voltage 11 kV on the crystal with 14,5х10,5х10 mm$^3$ surface area. The article describes the possibility to control the start of generation by introducing priming pulse. The results achieved are explained by the domain repolarization while heating the crystal and the appearance of electric field local strength. Bias and overcharge currents contribute to the appearance of electric strength, which stimulates breakdown and plasma formation. X-ray radiation appears both at the stage of discharge formation and during electron deceleration on gas and target material.

1. Present state of the problem
Lithium niobate is a promising material for creating compact solid-state x-ray emitters and electron accelerators based on pyroelectric effect [1-4]. However, insufficient radiation power restrains solid-state emitters from their practical application. Irradiating crystal with laser, increasing the number of elements and engraving allow to intensify the electric field formation on the crystal surface. In [2] we show the impact of plasma in the formation of emission from piezoelectric crystals. In this respect, the range of pressure 1-10 Pa is quite attractive, as it allows to realize high-voltage discharge and charge current gas amplification.

2. Problem solution technique
The solution of the radiation intensification problem is based on changing crystal parameters via doping and analyzing radiation generation processes from the point of view of plasma emission electronics. Doping allows to change crystal capacity and its resistance, and to activate junctions which stimulate excited atom migration. Heating the crystal leads to formation of electric field local strength areas as a result of domain vector polarization change. Breakdown between these areas becomes the source of plasma. Short-wave radiation (ultraviolet, x-ray) appear both from plasma and as a result of electron deceleration on gas or target material. This paper considers generation of electron stream in vacuum chamber at 1-10 Pa.
3. Process modeling

Figure 1 shows the model structure of the radiation generation. Crystal with radiation surface $S$ is placed at some distance $d$ from target $l$. To make it simple, let’s assume that crystal 2 is cylindrical and there is no electric field at the end faces. It’s supposed that changing the crystal temperature $T$ leads to the formation of local area 3 of electric field strength $E$ because of crystal domain repolarization. This causes gas ionization, breakdown and electron emission from discharge plasma. Figure 2 shows the assumed diagram of current and voltage time change.

![Figure 1](image1.png)  
**Figure 1.** Emitter model structure: 1 – anode; 2 – crystal; 3 – field strength area.

![Figure 2](image2.png)  
**Figure 2.** X-ray generation diagram: 1 – voltage characteristic; 2 – discharge current characteristic; 3 – X-ray pulse.

The process of x-ray radiation generation during crystal heating proceeds in several stages. At time $t_0$ the crystal is excited and the areas of electric field strength appear on it. At a specific distance, the field strength gets sufficient enough to ionize residual gas. This enables the formation of electron avalanche with the voltage $U_1$ (characteristic 1) and current (characteristic 2). The ions produce plasma cloud, which results in the increase of current and gap breakdown. Beginning with time $t_2$, when the current gets reasonably large, the plasma finite conductivity, restricted by charged particles mobility and deionization processes, starts to show. During the times $t_4$ from $t_1$ to $t_3$ in the area where there is both current and voltage, we can observe shock processes with generation of x-ray, ultraviolet and other radiations (characteristic 3). To implement electron emitter based on pyroelectric effect it’s essential to provide the current-voltage correspondence. The absence of current (idle stroke) or the absence of voltage (short circuit mode) indicates the non-operability of an electron emitter. The correspondence of current $i(t)$ and accelerating voltage $U$ at particle emission is set by the three-thirds power law [5]:

$$i(t) = 2.33 \cdot 10^{-6} U^{3/2}(t) \frac{S}{d}$$  \hspace{1cm} (1)

where $S$ is current take-off area; $d$ is the distance from emission zone to anode (target).

The charge presence is based on the Paschen Law, which correlates the ignition voltage $U_3$ and the product of pressure $P$ and distance between electrodes $d$:

$$(U_3 = f(Pd)).$$

The situation when the charge doesn’t ignite can be described by the inequality [6]:

$$\gamma(e^\alpha - 1) < 1,$$  \hspace{1cm} (2)

where $\gamma$ is the secondary ion-electron emission factor; $\alpha$ is Townsend ionization coefficient.

Electric field strength $E$ appearing as a result of pyroelectric effect determines the breakdown voltage $U$:

$$U = E \cdot d$$  \hspace{1cm} (3)
In the process of formation of electron avalanche, the shock ionization factor increases ($\alpha > 1$) and the conditions of self-maintained discharge are realized [7]:

$$\alpha d = \ln(1 + 1/\gamma)$$  \hspace{1cm} (4)

In this case electron current component $I_e$ can be found by [6]:

$$I_e = n_e V_e e S$$  \hspace{1cm} (5)

where $n_e$ is plasma concentration; $V_e$, $e$ are electron velocity and elementary charge respectively.

In the simplest case, electron velocity excluding gas encounter can be calculated from kinetic reasoning [8]:

$$I_e = n_e V_e e S$$  \hspace{1cm} (6)

Parameters modeling was conducted with MATHCAD software.

4. Sample preparation technology

The samples of lithium niobate were obtained by Czochralski method. While doping some crystals, $\text{Fe}_2\text{O}_3$ was added to the initial mix. The concentration of doping material was determined with interference [9] and SIMS [10, 11] techniques. To conduct research we used the crystals of different size with $Z$-oriented plane of polarization. The samples were subjected to microscopic study to analyze polishing accuracy, chip and scratch defects, and surface contaminants. Then, the samples were dried at 500K for 24 hours.

5. Experimental studies

The studies into electron emission from lithium niobate were carried out with the standard general-purpose exhaust cart VUP-5. Figure 3 shows the experiment schematic course, figure 4 demonstrates sample parameters.

Peltier element 1 of the type TEC1-12706 was connected to power supply unit HY3005-3. Crystal 2 was heated or cooled through copper plate. The temperature was measured with thermal converter attached to multimeter Protek 505. Anode 3 and electron collector 4 were arranged in the form of a Faraday pyramid at the distance of 3, 5, 10 mm from the crystal. Providing the pressure is ~1.33 Pa and heating the crystal proceeds at 10 - 20°/min, there appear surface charge areas on the crystal surface, which cause breakdown and plasma formation. Electron current flows towards anode 3. Current signal from resistor 8 falls on the oscilloscope C8-17 (5). The fragments from oscilloscope pictures are recorded onto the camcorder 6 and processed with PC 7. The resistor resistance ($R=10$ Ohm) was chosen in accordance with the conditions of minimal impact of thermal noise on the current.

Sample parameters: length, width, thickness, concentration
1 – 15 x 6 x 2 mm; $C_{\text{Fe}^2+} = 3.88 \cdot 10^{23}$ m$^{-3}$;  
2 – 18 x 21 x 3.5 mm; $C_{\text{Fe}^2+} = 3.01 \cdot 10^{23}$ m$^{-3}$;  
3 – 14.5 x 10 x 10.5 mm; $C_{\text{Fe}^2+} = 1.73 \cdot 10^{23}$ m$^{-3}$;  
4 – 10 x 4 x 4 mm undoped
detected at heating the crystal. The realizable breakdown voltage was estimated with the equation (1). The x-ray intensity was estimated by the blanket exposure of x-ray photographic film and compared with the negatives found in medicine. The pulse energy was estimated by the oscilloscope picture with the equation:

$$W = \int_0^t i(t) \cdot U(t) dt$$

6. Results and outputs

6.1. Undoped crystal (No 4).

Changing crystal temperature is followed by the appearance of voltage in it. This results in self-induced, spontaneous generation of electrons, as well as bias and overcharge currents, occurring at the frequency ~ 1 MHz. The largest intensity of spontaneous electron stream generation is observed at the pressures ~ 1 Pa and the temperatures 425-375 K. Increasing the pressure to 10 Pa results in a slight increase of current, which can be explained by the “gas amplification” at which ionization factor increases with the pressure rise. The interval between radiating emissions is 1-3 minutes. The crystal cooled down through its own heat capacity at 10°/min.

Figure 5 shows oscilloscope pictures of collector current at 1 Pa. Figure 6 shows the x-ray recorder.

**Figure 5.** Oscilloscope pictures of collector current at 1 Pa for undoped crystal.

**Figure 6.** The x-ray recorder on a photographic film.

The series of oscilloscope pictures prove the gas gap breakdown along the crystal surface. Oscillogram “downfall” near the maximum tells about current take-off. Design stress was 20-45 kV with the current up to 10 nA.

The intensity of film blanket exposure after tenfold radiation treatment in vacuum is 0.04 mz according to the results of photometry and referencing. Based on [8] the deceleration radiation intensity is directly proportional to the current, target material atomic number and the square of accelerating voltage. No more than 1 per cent of input voltage turns into x-ray radiation.

Managing the moment of generating radiation.

The migration of electric field strength area due to spontaneous local repolarization under temperature causes the instability of radiation generation moment. Distance d from the emission bound to accelerating electrode grows with the rise of accelerating voltage and reduces with the densification of extractable particles.

Introducing additional electrode into the crystal region allows for the preliminary pulse electric spark from capacitive discharge (1000 V, 5 mA, 50 ms). Plasma generation in the region of crystal local electric field enables to initiate the main discharge along the surface of piezoelectric followed by the current take-off.

Preliminary pulse allows to reduce the discharge time and to weaken electron stream intensity.
6.2. Iron doped crystals

Figure 7 shows oscilloscope pictures of signals obtained while heating doped crystal №1 to 90°C and cooling it to -20°C. The distance from the crystal to anode was 10 mm.

![Figure 7. a) Oscilloscope picture of the current at heating the crystal; 1mV/div=0,5 mA, t – 2μs/div b) oscilloscope picture of the current at cooling the crystal 1 mV/div =0,5 mA, t – 2 μs/div](image)

Increasing the temperature leads to increasing electric field strength and producing breakdown between crystal surface and anode. In pressures ~ 1Pa there is a possibility of a short non self-maintained glow discharge striking. Relatively permanent peak current intervals while heating can be explained by the gas gap breakdown between the areas of local strength. Current oscillations after the first breakdown result from the processes of domain recharge, deionization and recombination on the crystal surface.

Reducing the distance between the crystal and anode up to 3-5 mm results in transition of discharge into high voltage structure with higher breakdown voltages and more expressed current fluctuations. Current pulses at $P_d = 0.5 - 1$ correspond to voltages up to 5 kV. For copper anode ($\gamma = 7$) impact-ionization rate is $\alpha = (5 - 23)$, $U = 11$ kV, $I = 3$ mA. The obtained values of current and voltage correlate to the equation (1). Higher values of voltage pulses correspond to $P_d = 5 - 8$, however, under these circumstances the conditions for maximum values of current and voltage are instable. This agrees with [7] and is probably caused by the slope of the left branch of Paschen curve with respect to reference axis when one $P_d$ can have two appropriate ignition voltages.

Peak currents while heating and cooling the crystal are commeasurable. However, after several cycles of heating and cooling, discharge current at breakdown decreases fivefold or six fold. Changes in current and voltage result from sorption and desorption processes happening in the process of changing crystal temperature in the areas of fore vacuum pressure values.

Increasing the size of samples leads to increasing the current proportionally to crystal area. As a result, peak current value (3mA) was achieved with sample №3 having the size $14.5 \times 10.5 \times 10 \text{ mm}^3$ while heating it at 20°C/min from -10 to 107°C. Current pulses appeared with the interval 1-3 minutes at temperatures 17, 38, 56, 94, 98, 100, 105, 106, 107°C. Thus, iron-doped samples with $C_{Fe^2+}$ equal to $3.88 \times 10^{23}$ m$^{-3}$ allow to increase emission current to several milliamps. Crystal cascading results in increasing breakdown voltage, but it restrains crystal heating and cooling possibilities.

7. Conclusions

During the research, the new data on possibilities to generate electron stream in the process of heating and cooling the crystal at pressures 1-10 Pa were obtained. Iterative intervals on oscilloscope pictures show the gas gap breakdown rather than crystal surface breakdown. After the breakdown, bias and overcharge currents are realized. These currents maintain remaining residues of electron avalanche and prevent the discharge from instantaneous extinction. The studies [8-9, 12] prove the feasibility of the suggested model for generating electron and x-ray radiation. The well-defined regularity of
changes in radiation characteristics depending on the distance, crystal size, heating and cooling rates, and arranging system elements was established. Choosing doping material and its concentration allows for new prospects in developing solid-state optical electronic devices and x-ray equipment based on lithium niobate.

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