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Study of the pyroelectric luminescence of crystals

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Abstract

The pyroelectric luminescence accompanying a change in the temperature of a pyroelectric crystal is associated with electric fields which appear in the crystal as a result of the pyroelectric effect. The surface charge density of pyroelectric crystals increases gradually with temperature and heating or cooling time, in which the increase is observed over a temperature range correlated to the time elapsed between two successive light pulses. Following the charging and discharging of the crystal surface, expression is derived for temperature and time dependant pyroelectric field and pyroelectric luminescence, in which a good agreement is found between the theoretical and experimental results.
Keywords: pyroelectric luminescence, crystal surface.

1. INTRODUCTION

Pyroelectric luminescence (PEL) is the phenomenon of light emission produced during heating or cooling of solids having pyroelectric behavior [1]. The pyroelectric luminescence accompanying a change in the temperature of a pyroactive crystal is associated with electric fields which appear in the crystal as a result of the pyroelectric effect, and has already been observed in many crystals exhibiting the pyroelectric effect, such as BaB_2O_4 [2], $LiNaSO_4$:Eu [3,4], $LiNbO_3$, tourmaline [5], proustite, pyraagyrite [6], lithium metagermanate [7], BeO [8], LiB_4O_7 [9] and LiB_3O_5 [10], etc, The mechanism responsible for this pyroelectric luminescence is assumed to differ for different crystals, ranging from a pure electric discharge at the surface of the crystal to the combination of surface breakdown, internal breakdown, and self-radiation of crystal. The present paper reports the theory of PEL and makes a comparison between the theoretical and experimental results.

2. THEORY

Let us consider an ideal pyroelectric single crystal placed in a continuous gas medium of dielectric permittivity ϵ_g. The change in temperature modifies the positions of the atoms slightly within the crystal structure, such that the polarization of the material changes. This polarization is mathematically given by

$$P = n\mu \quad (1)$$

where n is the number of molecules per unit volume and μ is the induced microscopic dipole moment. We can express this spontaneous polarization by the microscopic dipole moment μ of the crystal with the integral running over its volume as

$$P = \int_V \frac{\mu dV}{v} \quad (2)$$

The temperature induced change in the spontaneous polarization, accompanied by the spontaneous electric field growing along the pyroelectric axis of the crystal can be expressed as

$$E_S = \frac{P}{\epsilon_0 \epsilon} \quad (3)$$

Using Eq. (2) we can express the spontaneous electric field as

$$E_S = (\epsilon_0 \epsilon V)^{-1} \int_V \frac{\mu dV}{v} \quad (4)$$

where ϵ is the dielectric permittivity of the crystal and ϵ_0 is the dielectric permittivity of the free space.

The field in the crystal near edge region of the gas is related with E_S through the simple relation

$$E_g = \frac{\epsilon}{\epsilon_g} E_S \quad (5)$$

or, $$E_g = \frac{\sigma_s}{\epsilon_0 \epsilon_g} \quad (6)$$

where σ_s is the surface charge density.

When E_g exceeds E_S, probability of the electrical breakdown of the gas becomes greater than that of crystal itself.

As PEL directly represents pyroelectric properties of the crystal, the pyroelectric coefficient after uniform heating or cooling of the crystal is given by

$$p(T) = \epsilon_0 \epsilon \frac{dE_S}{dT} \quad (7)$$

The field E in the crystal may reach the value of the breakdown field strength E_b only if the charge generation rate exceeds the rate of its compensation. In this case, the balance equation can be written as

$$\epsilon_0\epsilon(T)\frac{dE_s}{dT} = p(T)\frac{dT}{dt} - \sigma(T)E(T) \quad (8)$$

If β is the uniform heating or cooling rate of the crystal, then the rise of temperature T can be expressed as

$$T = T_0 + \beta t \quad (9)$$

where T_0 is the temperature at $t = 0$, $\beta > 0$ for heating and $\beta < 0$ for cooling and $\frac{dT}{dt} = \beta$.

Rearranging Eq.(8), we get

$$\frac{dE(T)}{dT} = \frac{p(T)}{\epsilon_0\epsilon(T)}\frac{dT}{dt} - \frac{\sigma(T)}{\epsilon_0\epsilon(T)}E(T)$$

or, $$dE(T) = \frac{p(T)}{\epsilon_0\epsilon(T)}\beta dt - \frac{\sigma(T)}{\epsilon_0\epsilon(T)}E(T)dt \quad (10)$$

We can write Eq.(10) as

$$dE(T) = \alpha dt - \gamma E(T)dt$$

or, $$dE(T) = [\alpha - \gamma E(T)]dt \quad (11)$$

where $\alpha = \frac{p(T)}{\epsilon_0\epsilon(T)}\beta$ and $\gamma = \frac{\sigma(T)}{\epsilon_0\epsilon(T)}$.

For a limited time interval α and β may be considered as constants, and thus, after solving Eq.(11), we get

$$E(T) = \frac{\alpha}{\gamma}(1 - e^{-\gamma t}) + e^{-\gamma t}E(T_0) \quad (12)$$

As $E(T_0) = 0$, at $T = T_0$. Eq.(12) can be reduced as

$$E(T) = \frac{\alpha}{\gamma}(1 - e^{-\gamma t}) \quad (13)$$

After attending $E(T) = \frac{\alpha}{\gamma}$, the gas discharge occurs and $E(T)$ becomes zero, and subsequently, the recharging of the surface of crystals with increasing temperature takes place. Thus, we can express Eq.(13) as

$$E(T) = \frac{\alpha}{\gamma}[1 - e^{-\gamma t}] + \frac{\alpha}{\gamma}[1 - e^{-\gamma(t+t_0)}] + - -$$
$$+ \frac{\alpha}{\gamma}[1 - e^{-\gamma(t+nt_0)}] \quad (14)$$

where t_0 is the time taken for charging the crystal for discharge to occur and n is the number of PEL flashes.

3. EXPERIMENTAL SUPPORT TO THE PROPOSED THEORY

Fig.1 shows that the surface charge density increases gradually with temperature and time, whereby the increase being observed over a temperature range correlated to the elapsed between two successive light pulses. During this time the electric field is building up to the threshold value of the breakdown field needed to produce the luminescence. The rise time of this charge is followed by its fast decay of duration corresponding to the time of discharge. This finding supports Eq.(14).

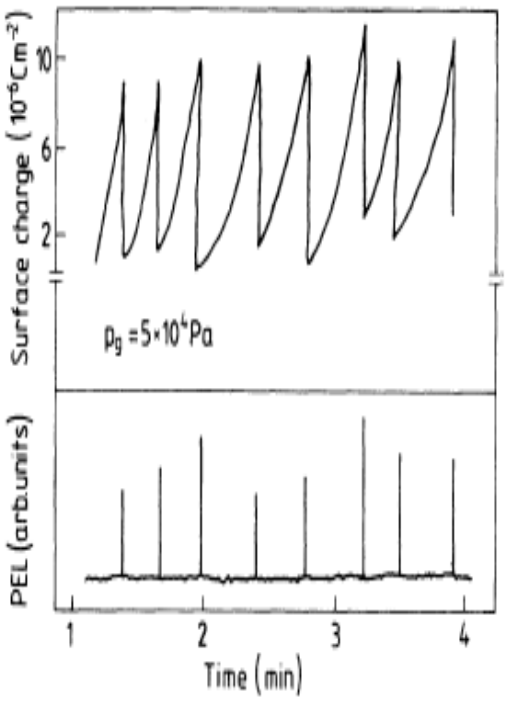

Fig. 1 Bottom: Measured intensity of light emitted by a crystal of NIPC when cooled at the rate β = - 0.083 Ks^{-1} starting at 270 K. Top: Simultaneous measurement of the surface density of the charge created on the crystal upper surface perpendicular to the pyroelectric axis of the crystal(after Kalinowski et al., ref.[1]).

Fig.2 shows the temperature dependence of the integrated emission of the BaB_2O_4 single crystal during cooling (curve 1) and heating (curve 2). It can be seen clearly that the luminescence of the BaB_2O_4 single crystal, in the form of isolated flashes, is recorded over the entire range of temperature (80-400 K). The amplitude and repetition frequency of the light flashes vary depending on the rate of change in temperature and the ambient gas pressure, and decrease slightly at high temperature. When the temperature stabilizes after a change, the light flashes cease. The characteristics of BaB_2O_4 luminescence with varying temperature indicate that the observed luminescence has a pyroelectric charac- pyroelectroluminescence. Adamiv et al. (reference [2]) have reported that, when a β-BaB_2O_4 crystal heated or cooled at a fixed heating rate or cooling rate, Then PEL flashes appear with increase in temperature of the crystal. Such fact indicated by Eq.(14).

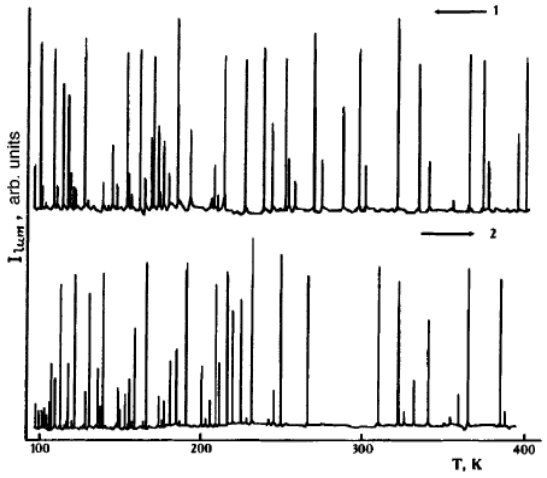

Fig.2 Temperature curves of I_{lum} for a β-BaB_2O_4 crystal during cooling (curve 1) and heating (curve 2). The direction of change in temperature is indicated by the arrows (after Adamiv et al., ref. [2]).

4. CONCLUSION

The surface charge density of pyroelectric crystals increases gradually with temperature and heating or cooling time, in which the increase is observed over a temperature range correlated to the time elapsed between two successive light pulses. Following the charging and discharging of the crystal surface, expression is derived for temperature and time dependant pyroelectric field and pyroelectric luminescence, in which a good agreement is found between the theoretical and experimental results.

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