the lower temperature peaks tend to decrease with increasing irradiation dosage although the amount of complex centers increases with dosage. This appears to me to be in contradiction with your interpretation.

Ueta, M.: As I showed in the first slide, the thermoluminescence intensity depends much on the crystal history and changes to crystals to crystals cleaved, even in the crystals from the same ingot. Therefore the quantitative comparison of glow intensity with optical density of complex centers should be made very carefully.

Klick, C. C.: It should be interesting to know whether the emission spectrum in any of the thermal glow peaks is identical with that seen by Onaka, Fujita and Fukuda* reported in their paper on irradiation in the α center. If such an identification could bemade it might allow the assignment of one of the peaks as due to the recombination of an exciton at an isolated vacancy.

* *Proc. Int. Conf. Cryst. Latt. Def. (1962)*: J. Phys. Soc. Japan **18** Suppl. II (1963) 263.

PROCEEDINGS OF THE INTERNATIONAL CONFERENCE ON CRYSTAL LATTICE DEFECTS, 1962, CONFERENCE JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 18, SUPPLEMENT II, 1963

Electron Conduction and Breakdown in Gamma Ray Irradiated KCl Crystals

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Pulse F band photoconduction upto prebreakdown range and dielectric breakdown field of γ irradiated KCl crystals were investigated. From the electric field saturation of pulse photocurrent, mobility $\times$ life time ($\mu\tau$) and relative value of quantum yield for photoconduction (η) were obtained at various temperature and F center density. Apparently η increases at about -60°C with increasing temperature. $\mu\tau$, however, decreases. Current multiplication possibly due to impact ionization of F center were found only just below the breakdwon field. In nearly perfect KCl, temperature dependence of breakdown field well coincides with the theory which takes into account the electron interaction with polar optical phonon. γ ray introduced defects increases low temperature pulse breakdown field possibly through inelastic impurity scattering and also increases d.c. breakdown field around room temperature due to the space charge effect of the trapped electrons.

1. Introduction

von Hippel's early experiments[1] on dielectric breakdown of alkali halide crystals revealed the nature of lower temperature breakdown to be the results of electron avalanche with penetrating intuition based on the effects of crystal imperfections, and have been followed by a series of theoretical formulations by many workers[2]. The efforts to compare these theoretical value with experimental data have confronted various difficulties partly due to improper experimental data suffering from secondary factors such as dislocations and vacancies. We have tried[3] to get nearly intrinsic breakdown data in KCl crystal to compare with the theory and to clarify the effect of point defects on the dielectric breakdown. The conventional measurement of the breakdown field only can not reveal the mechanism without knowing the behaviours of hot electrons at prebreakdown range. To this purpose F band pulse photoconduction upto prebreakdown range and breakdown field were measured simultaneously for KCl crystal in which controllable amount of imperfections had been introduced by γ irradiation.

2. Experimentals and Discussions

KCl single crystals carefully grown by Kyropoulos method after recrystallization of three times were well annealed for 10 hr at 600°C, then cooled to room temperature in 20 hr. These crystals were cleaved, shaped to $10\times10\times0.3$ mm³, reannealed, and furnished with vacuum evaporated gold electrode of diffused edge to one side of the sample (conduction sample) or to the both sides (breakdown sample). Known amounts of γ ray from Co^{60} were irradiated on these samples at room temperature to introduce lattice defects with the dose rate of 3×10^5r/hr. Optical absorption of the irradiated KCl in Fig. 1 show predominant F band absorption. This curve with Smakula's formula allowed us to estimate F center density.

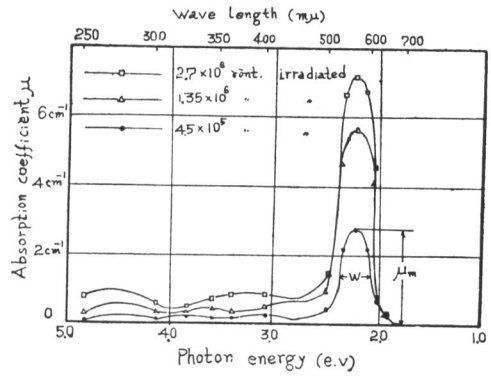

Fig. 1.

(a) F band (550mμ) pulse photoconduction

Firstly the peak value of the photocurrent I in irradiated KCl illuminated with a 550mμ

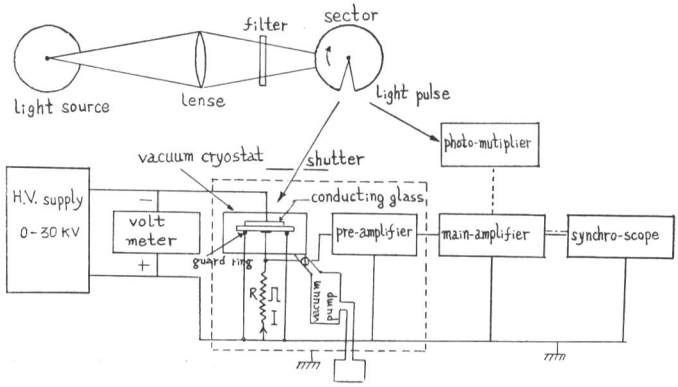

Fig. 2.

light pulse of 500μs duration were obtained at various d.c. applied field E and temperature with the apparatus shown in Fig. 2. Exposure to the light pulse of constant intensity (50 lux: 2×10^{13} photon/cm² sec) was limited to the minimum time required for photographing synchroscope figure by the shutter in order to prevent space charge effects. Injected photon into the effective region of the crystal per pulse was ca. 10^9. KCl samples were inserted into a vacuum cryostat system with conducting glass cathode through which the light pulse was applied. In the case of uniform release, the photocurrent I at an applied d.c. field E can be given by the well known Schubweg formula.

$$I=e\eta N_0\frac{w}{l}\left[1-\frac{w}{l}[1-\exp(-l/w)]\right] \qquad (1)$$

$$w=\mu E\tau\cdots\cdot\text{Schubweg}$$

where η, N_0, l, μ and τ means quantum yield, absorbed photon number per unit time, sample

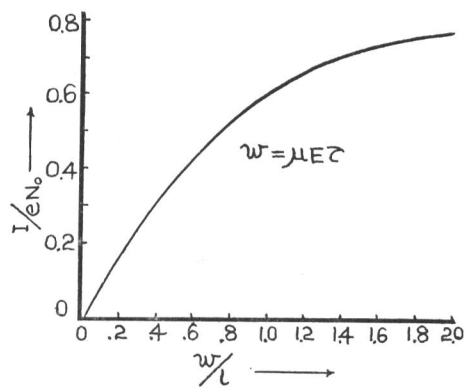

Fig. 3.

thickness, mobility and life time respectively. Eq. (1) gives Schubweg saturation curve of the photocurrent I *vs.* w/l or applied field E as shown in Fig. 3. Moreover, it gives

$$\left.\begin{array}{l}\text{the low field conductivity}\\ \qquad \lim_{E\to 0}(I/E)=eN_0\eta\mu\tau/l\\ \text{and the saturation current}\\ \qquad \lim_{E\to\infty} I=eN_0\eta/2.\end{array}\right\} \quad (2)$$

Experimental saturation curves of the peak photocurrent under a constant pulse light intensity at various temperature and F center density N_F are shown in Figs. 4 and 5. The reproducibility of the curves was satisfactory due to slight space charge effect under a limited number of photons. By comparing the experimental curve in Fig. 3 and Eqs. (1) and (2), η and $\mu\tau$ could be determined separately as shown in Fig. 6 and Table I. Strictly speaking, however, the value of η should be taken to be in arbitrary unit since the exact measurement of incident photon number N_0 was not done. The value of η at room temperature seems to be almost equal to unity. Above mentioned Schubweg analysis is not justified at lower temperature (−160°C) where Zener emission from excited F state makes η strongly field dependent as was shown by Lüty's curve[9] in Fig. 5. $\mu\tau$ decreases with increasing temperature according to decrease in μ, and τ is inversely proportional to F center density, being rather insensitive to the temperature as

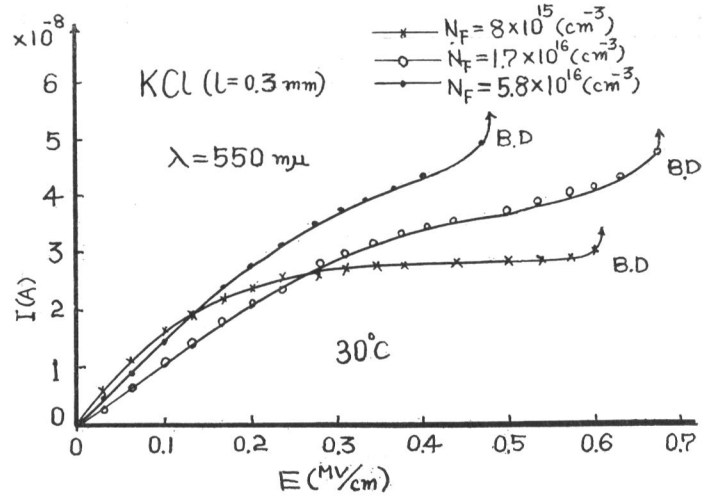

Fig. 4.

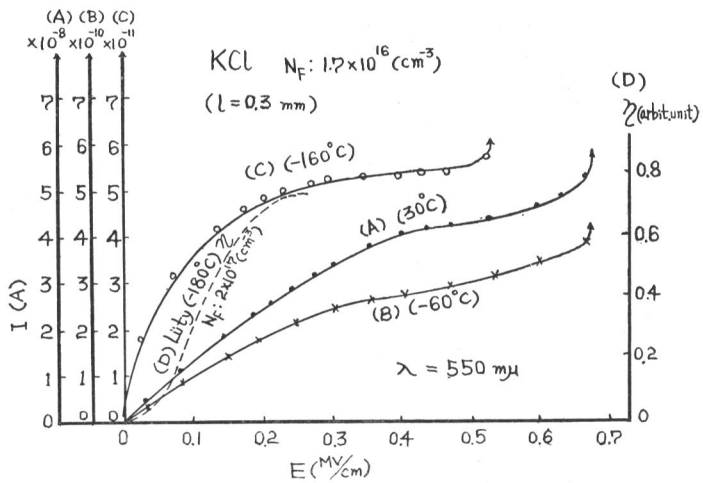

Fig. 5.

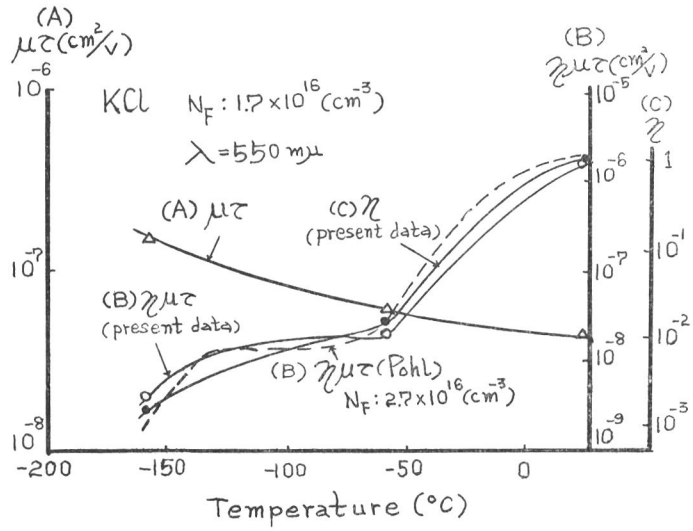

Fig. 6.

Table I.

temperature	$\mu\tau$(cm²/volt)	η	τ(sec)	E_{Bd}(MV/cm)	d_B(cm)
30°C	N_F(cm^{-3}):1.7×10^{16} 0.8×10^{-7}	1	0.6×10^{-8}	0.67	0.044
	N_F(cm^{-3}): 8×10^{15} 1.5×10^{-7}		1.5×10^{-8}	0.61	0.101
−60°C	N_F(cm^{-3}):1.7×10^{16} 0.80×10^{-7}	0.7×10^{-2}	0.5×10^{-8}	0.67	0.054
−160°C	N_F(cm^{-3}):1.7×10^{16} 1.8×10^{-7}	0.98×10^{-3}	0.7×10^{-8}	0.59	0.106

shown in Table I. As seen in Fig. 6, $\eta\mu\tau$ and η have similar temperature dependence and the increase of pulse photocurrent above −60°C seems to be not due to increase in τ by the thermal release of F' center, but due to increase in apparent η. The thermal release time of F' center at room temperature obtained from Gericke's formula[6] supports this view, being much larger than our pulse duration 500μs. Although the detailed mechanism of the apparent η increase above −60°C is unknown at this stage of the experiment, the increase in the escape of photoelectrons from coagulated F center around dislocations with much shorter life time, due to close neighbouring vacancies and also possibly due to lower conduction band edge, is speculated to be the case. The fact that superposition of 750mμ (F' band) light enhanced F band photocurrent above −60°C is consistent with this speculation. At prebreakdown field very near to final breakdown, small charge multiplication sets in as seen in Figs. 4 and 5, being possibly

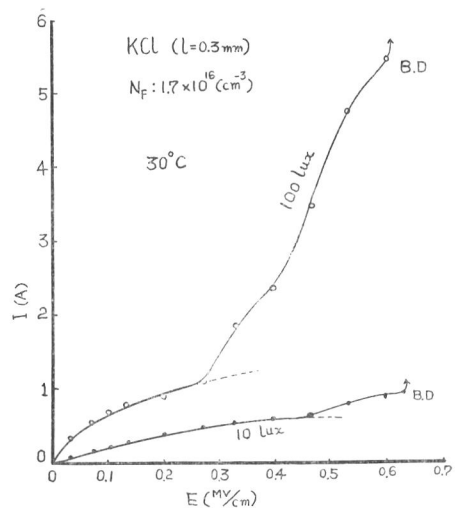

Fig. 7.

due to impact ionization of F centers. Accordingly, electrons in ionic crystal does not become markedly hot (several eV) until just below breakdown field due to strong interaction with polar mode lattice vibration, thus supporting low energy criterion for breakdown. These multiplication was apparently enhanced with increasing light intensity as shown in Fig. 7, being possibly due to cathodic field enhancement through positive space charge of photoionized F centers which results in lower breakdown strength. Smaller value of apparent η at lower temperature in Table I than that from optical data should be noted.

(*b*) *Breakdown electric field*

The impact ionization breakdown theory, taking into account the interaction of electrons with longitudinal optical lattice vibration, predicts that the temperature dependence of the breakdown field $E_B(T)$ at a temperature T°K is given by

$$E_B(T)=E_B(0)(2n+1)^{1/2} \qquad (3)$$

where

$$n=\left[\exp\left(\frac{\hbar\omega_t(\varepsilon_0/\varepsilon_\infty)^{1/2}}{kT}\right)-1\right]^{-1}$$

is the mean number of the quantum of lattice vibration. ε_0 and ε_∞, ω_t and k mean static and optical dielectric constant, angular frequency for transverse optical mode of vibration, and Boltzmann constant respectively. The comparison of this theoretical value (full line) with the experimental pulse ($0.1\times100\mu$s) breakdown field (circle) of the carefully prepared unirradiated KCl are given in Fig. 8, which shows good agreement to each other providing that E (93°K)/E (0)=1.02, and taking $\hbar\omega_t(\varepsilon_0/\varepsilon_\infty)^{1/2}$ to be 0.028 eV. The deviation at high temperature might be due to the change in breakdown mechanism. The impulse breakdown field does not depend on sample thickness in the range 0.2mm-0.6mm, proving to be intrinsic. With increasing F center density or the dosage of preirradiation, d.c. and impulse breakdown field in the dark at room temperature varies as shown by the full line in Fig. 9. Although d.c. breakdown field shows predominant increase from 0.5 MV/cm to 1 MV/cm with increasing F center density, the impulse value shows slight change. Accordingly, abnormal increase of d.c. breakdown field above room temperature may be due to the negative space charge effect of trapped electrons which will be discussed later. The illumination with F band light during d.c. breakdown lowers the breakdown field markedly as shown by dotted line in Fig. 9. This phenomenon, as was published elsewhere[8],

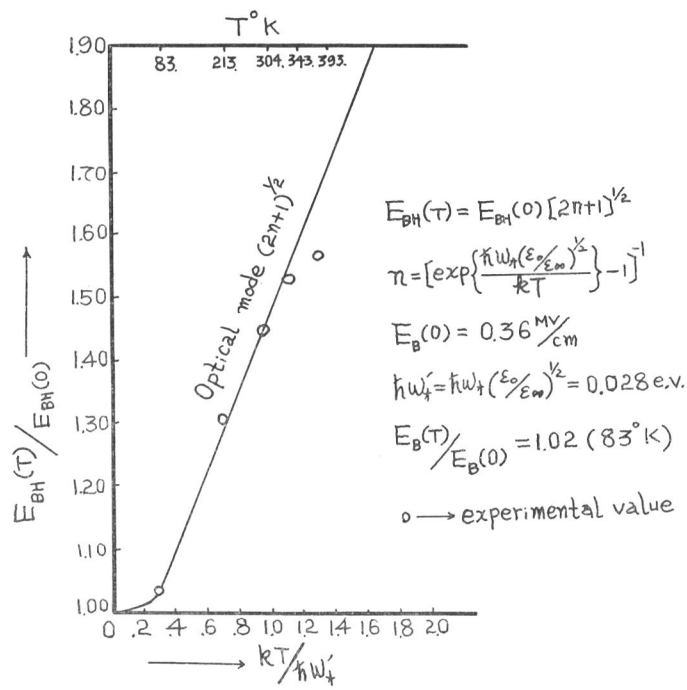

Fig. 8.

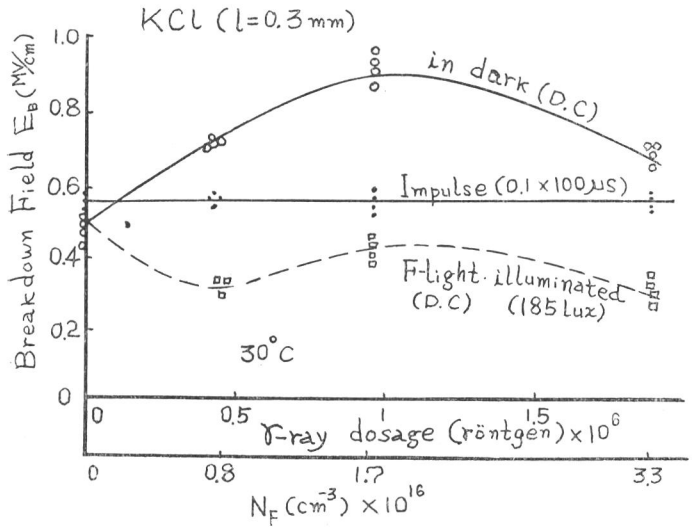

Fig. 9.

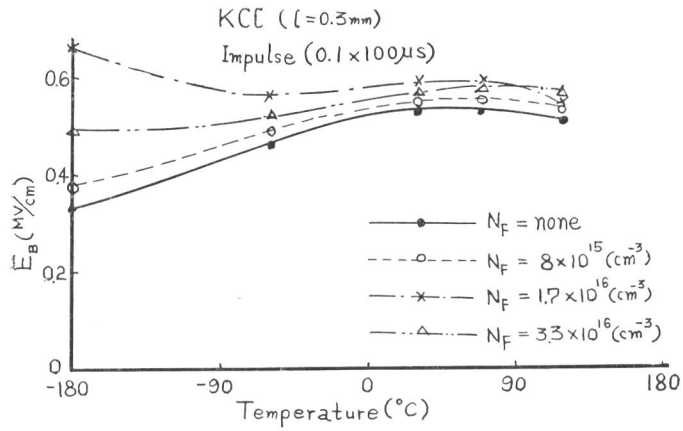

Fig. 10 (a).

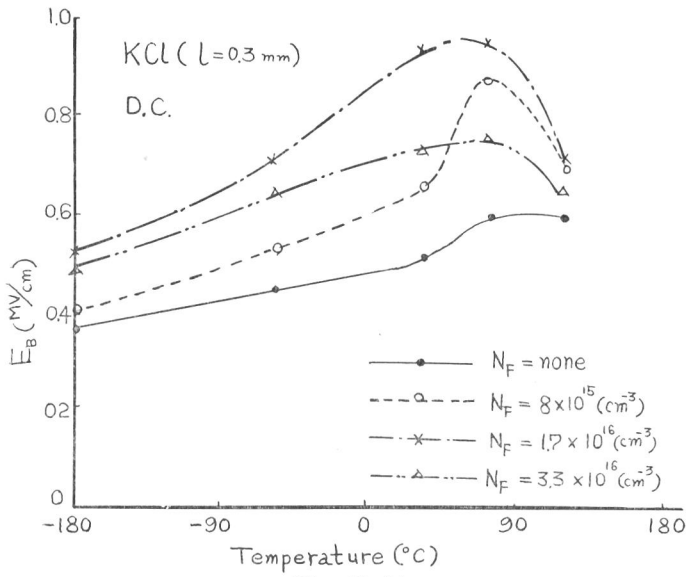

Fig. 10 (b).

seems to be due to cathodic field enhancement through the formation of positive space charge due to photoionized F center.

The temperature dependence of the impulse breakdown field become flat with increasing F center density as shown in Fig. 10 (a), this is possibly due to inelastic impurity scattering of F center predominating at lower temperature. The temperature dependence of d.c. breakdown field, on the other hand, shows abnormally steep rise above 0°C as shown in Fig. 10 (b). Since this abnormal rise is not seen in the impulse value, we may consider that this effect is due to electronic space charge at vacancies generated with γ ray by the following mechanism. At lower temperature, since Schubweg d_B under breakdown field E_B in Table I is larger than the sample thickness 0.3 mm, field distortion due to trapped electron may be absent. Schubweg d_B decreases with increasing temperatue and finally approaches to the sample thickness. Then the negative space charge formation of trapped electrons may distort field distribution and raise the d.c. breakdown field through suppression of electron emission from cathode. The decrease in breakdown field at much larger F center density in Fig. 9 may be due to predominant impurity electron avalanche through impact ionization of F center. Tentative comparison of the experimental breakdown field at room temperature with the theoretical value, taking effective mass equal to free electron mass, are shown in Table II. Experimental value lies between high energy and low energy criterion value.

Table II.

von Hippel's field E_{BH}(MV/cm)	Seitz's field E_b(MV/cm)	Fröhlich's field E_{BF}(MV/cm)	Measured field E_B(MV/cm)
1.3	0.4	0.25	0.5

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