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SPACE – CHARGE – LIMITED CURRENT AND COLOUR CENTER FORMATION UNDER A SPHERICAL CONTACT ON KI SINGLE CRYSTAL

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Electron injection under different fields has been made in a spherical heterogeneous contact at 723 K in KI crystal. The current and colouration with time is discussed to justify the existence of space-charge-limited state.

1. *Introduction*

Space charge appears in a variety of situations in amorphous materials [1,2], semiconductors [3,4] and insulators [5-8]. In measurements of electrical conductivity in insulating solids at elevated temperature and applied field a transient current response has been observed [9]. Such response is characterised by the band structure [10], impurity and defect concentration of the crystal developed under heat treatment.

Montojo et al. [11] observed a characteristic growth of response in alkali halides and explained it in terms of the existence of the space charge within the bulk material in various zones. The present paper deals with space-charge-limited (SCL) current through electrolytes like single crystals of alkali halides due to a spherical contact. Here the current is controlled not only by the electron injecting electrode but also by the mobile carrier found within the space inside the material. Hence, this can be regarded as a bulk phenomena. The emitted electrons have a distribution of energies, while the crystal has traps of various distributions. Once the injecting contact can provide a reservoir of carriers, the current is controlled by the quality of trap distribution within the crystal.

2. *Experimental*

Pure KI powder (obtained from E-Merck) was heated at 873 K in a quartz ampule for several hours in order to eliminate posssible impurities. Single crystals of KI has been grown from melt by Kyropoulos method. A cleaved block along the <100> direction of dimensions 1 cm × 0.4 cm × 0.35 cm was taken from the central portion of the grown crystal and one face (0.40 cm × 0.35 cm) was rounded and water polished, making the radius of curvature the same as that of a platinum anode. The crystal is bound by a pointed brass cathode and a platinum anode into which the rounded portion of the crystal fitted perfectly (Fig. 1).

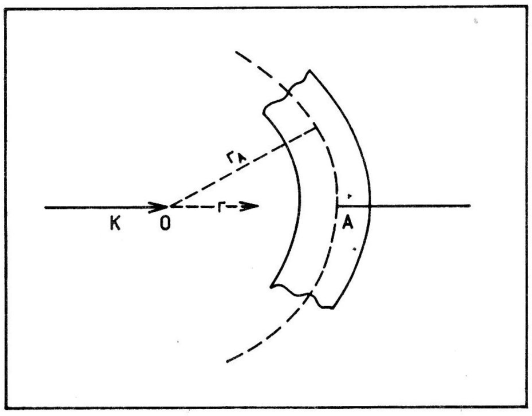

Fig. 1. Pointed cathode (K) and spherical anode (A) geometry.

The system was inserted in a furnace. The experimental details have been described elsewhere [12]. The injection was perfomed at 723 K under different fields between 75 and 200 V cm^{-1} separately. While shinning a wavelength of 685 nm, which is the maximum F-absorption light for KI, over a small portion of the crystal very near to cathode, using an Oriel grating monochromator, the growth of colouration and injection current as a function of time have been recorded simultaneously with the help of a Bausch and Lomb series 5000 strip chart two-pen recorder with pen speed 5 cm min^{-1} at each injection voltage. The injection procedure was stopped just before the advent of the third zone [13]. From the recorded data the graph shown in Fig. 2 was made displaying the dependence of colouration (optical density) and injection current on the injection fields.

3. Results and discussion

Optical density (O.D.) vs. current (I) graph shows a dependence of type O.D. $=a\ (I)^b$, where the arbitrary parameters a and b are evaluated by the method of averages. In order to determine the relationship between I and V, we determine values of current from each curve of Fig. 2 for a fixed value of O.D. (dashed line). A plot of $\log I$ vs. $\log V$ produces a straight line whose slope is found to be approximately equal to 2, employing the appropriate curve fitting technique. So, it appears that the colouration zone, where the current is proportional to the square of the voltage, confirms that we are in the SCL region [8].

Let us examine analytically the relationship, by considering single carrier injection from curved contacting electrodes.

The behaviour of injection in a solid, with a concentric spherical configuration is directed by the current flow equation given by,

$$I = e\mu_n nF(4\pi r^2)$$

and the corresponding Poisson's equation

$$1\frac{1}{r^2}\frac{d}{dr}(r^2F) = \frac{e}{\varepsilon}(n+n_t) \tag{1}$$

where n, n_t are, respectively, the density of injected free and trapped electrons, and F is the electric field in the crystal.

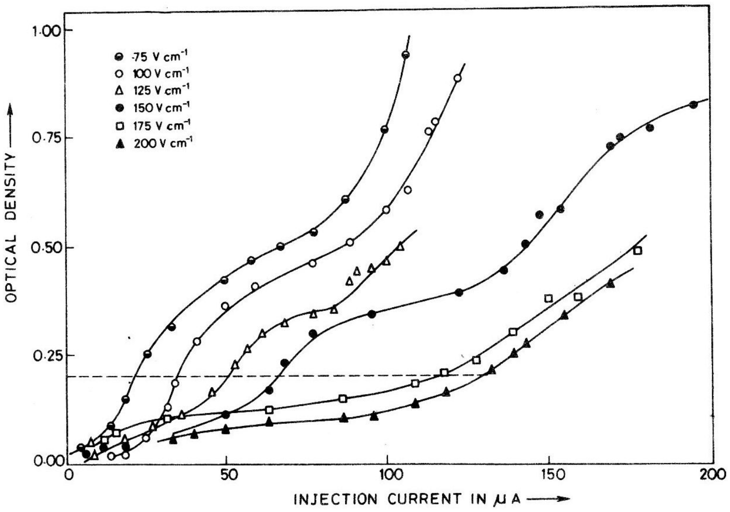

Fig. 2. Space-charge-limited current vs. colouration (O.D.) at 723 K *under different injection fields in* KI *crystal.*

We take

$$n = N_c \exp\left[\frac{(E_{Fn} - E_c)}{k_B T}\right]$$

$$2 \qquad n_t = \int\limits_{E_1}^{E_2} N_t(E) S(r) f(E) \mathrm{d}E \tag{2}$$

$$3 f(E) = \frac{1}{1 + g_n^{-1} \exp\left[\frac{(E - E_{Fn})}{k_B T}\right]} \,, \tag{3}$$

where:
N_c is the effective density of states in conduction band,
E_{Fn} the quasi Fermi level of electrons,
E_c the energy level of conduction band,
$f(E)$ the Fermi-Dirac distribution function,
g_n the degeneracy factor of trapped states for electrons,
$N_t(E)$ and $S(r)$ energy and spatial distribution functions of traps and E_1, E_2 are the lower and upper energy limits of trapping levels.

However, for shallow traps confined to a discrete energy level,

$$4 N_t(E) = H_e \delta(E - E_t) \tag{4}$$

where H_e is the trap density and E_t the trapped energy level below the edge of the conduction band.

On substituting Eqs. (3) and (4) in equation (2) and by taking $S(r) = 1$ for uniform distribution of trap density, we obtain

$$n_t = \frac{\int\limits_{E_1}^{E_2} H_e \delta(E - E_t) \mathrm{d}E}{1 + g_n^{-1} \exp\left[\frac{(E - E_{Fn})}{k_B T}\right]} \,.$$

Since the single discrete trap levels are fully occupied

$$\begin{aligned} n_t &= \frac{H_e}{1 + g_n^{-1} \exp\left[\frac{(E_t - E_{Fn})}{k_B T}\right]} \\ 5 \quad &= \frac{H_e}{1 + g_n^{-1} \exp\left(\frac{E_t}{k_B T}\right) \frac{N_c}{n} \exp\left(-\frac{E_c}{k_B T}\right)} \qquad (5) \\ &= \frac{n H_e}{n + \Theta_e H_e} \end{aligned}$$

where

$$\Theta_e = \frac{N_c}{g_n H_e} \exp\left[-\frac{(E_c - E_t)}{k_B T}\right] .$$

In case of shallow traps,

$$\frac{N_c}{g_n} \exp\left[-\frac{(E_c - E_t)}{k_B T}\right] \gg n$$

Eq. (5) takes the form

$$6n_t = \frac{n}{\Theta_e} . \tag{6}$$

Solving the Poisson's equation (1) using Eq. (6) and with the application of boundary conditions for spherical heterogeneous contact, we arrive at

$$I = \frac{3}{2}\pi t \mu_n \left(1 + \frac{1}{\Theta_e}\right) \frac{V^2}{r_A}$$

where the applied voltage $V = \int\limits_0^{r_A} F(r)\mathrm{d}r$ in the geometry of Fig. 1. Thus,

$$I \sim V^2 .$$

The situation is equivalent to the case of space charge in a vacuum diode but with a distinct difference in action. The analytic results corroborate with the experimental results and thus equally support the existence of such physical situation of a SCL current.

The growth of colour center develops at a rapid rate during SCL current condition and we have restricted the injection operation just before the advent of the third stage in a similar manner as conducted in our previous experiment [13]. In the present case both the injection current and the colouration are time dependent at constant field and temperature.

It is evident from Fig. 2 that for a particular field after a region of sharp rise of O.D. with current the curve flattens, indicating that although the current increases, colouration does not increase appreciably. Finally, there occurs again a steep rise of colouration with current. This may appear to be surprising but in fact it should be the case. The F-light shines over a very small region of the crystal and initially the colour center starts to move in a planar region. As time progresses, the multipronged colour centers occupy the bulk of the crystal throwing away the rest of the electrons to the anode by means of trapping, detrapping or directly. Consequently the colouration in this illuminated region lags with current.

Again to study the effect of field on colouration growth from Fig. 2 it is seen that for a particular current as field increases O.D. decreases. It has been observed that the efficiency of F-center formation [13,14] is a function of temperature and injection field. As a matter of fact all the electrons do not contribute to the colour center formation. Only a limited number of electrons are effectively trapped. With the increase of the field the probability of detrapping is significantly higher unless prolonged time of injection is being maintained leading to decrease in O.D. with field. To examine this further, we plot the rate of growth of O.D. with time in Fig. 3.

Figure 3 shows sudden rise and then the rate falls again and continues almost in a random manner and finally we see a steep rise of the rate. The rate of growth for application of a field between 75 – 150 $\mathrm{V\,cm^{-1}}$ suggests more or less the same pattern, but above 150 $\mathrm{V\,cm^{-1}}$ (say at 175 $\mathrm{V\,cm^{-1}}$) the situation seems to be different, when the colour centers grow almost in a uniform manner. Injection performed above 150 $\mathrm{V\,cm^{-1}}$ (apparently a critical voltage at temperature of injection 723 K), the carrier electrons accelerate rapidly from the cathode to anode in a rectilinear fashion due to a smaller number of electrons being trapped in voids. It is basically surface phenomenon. Around 175 $\mathrm{V\,cm^{-1}}$, though for a short while in the beginning, the tendency of trapping in a rectilinear path exists but relinquishes to the extent of being trapped in a greater number throughout the crystal leading to bulk phenomena. This is visible in the graph which shows a broad peak followed by random nature of colouration characterising bulk phenomena. Thus, we find that after the contact is being formed the SCL current phenomena prevail with characteristic growth of colouration.

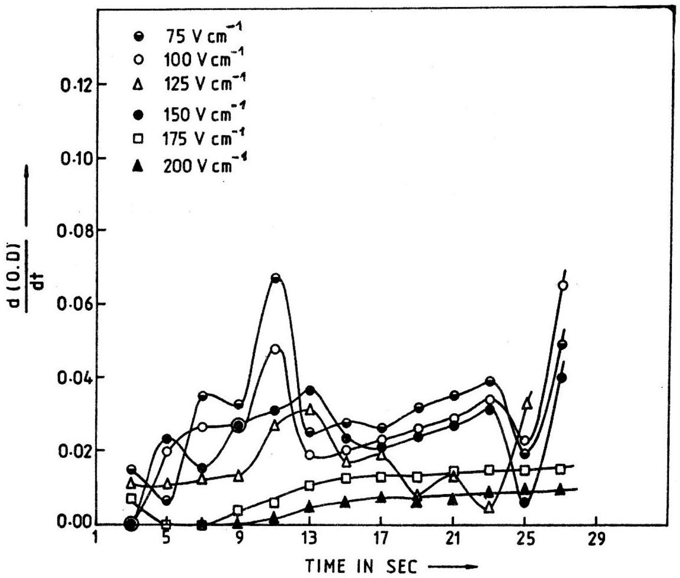

Fig. 3. Rate of colouration growth $\frac{\mathrm{d}}{\mathrm{dt}}$*(O.D.) vs. time for different injection fields.*

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STRUJA OGRANIČENA PROSTORNIM NABOJEM I STVARANJE CENTARA BOJE U KRISTALU KI S KRUŽNIM KONTAKTOM

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U KI kristal s heterogenim kružnim kontaktom injektirani su elektroni uz razne vrijednosti električnog polja. Raspravlja se ovisnost struje i obojenosti o vremenu kako bi se dokazalo postojanje stanja ograničenog prostornim nabojem.