JOURNAL OF OPTOELECTRONICS AND ADVANCED MATERIALS Vol. 12, No. 9, September 2010, p. 1840 - 1851

A new theory of the photoplastic effect in coloured alkali halide crystals

S. TIWARI[a], A. S. GOUR, V. K. JOGI[a], B. P. CHANDRA[b]
School of Studies in Physics, Pt.Ravishankar Shukla University, Raipur 492010
[a]School of Studies in Electronics, Pt.Ravishankar Shukla University, Raipur 492010
[b]Disha Institute of Management and Technology, Raipur 492101, (C.G.)

A new theory of the photoplastic effect (PPE) in colored alkali halide crystal, which is based on the interaction between moving dislocation and photo-obstacles has been formulated. When a colored alkali halide crystal is illuminated by light with certain wavelength during its plastic deformation, the increase or decrease in flow stress occurs because the photo-ionized electrons from F-centers influence the motion of deformation bleaching and photo-ionized electrons from F-centers influence the motion of deformation bleaching and interaction of moving dislocations with F-centers, expressions are derived for the PPE, like temporal , deformation, strain-rate, spectral, light intensity, coloration, dimension and thermal characteristics of PPE. A good agreement is found between the theoretical and experimental results. It is shown that the PPE provides a sensitive tool for determining the lifetime of dislocation-captured F-centers in crystals. It is shown that in certain crystals the radius of interaction between F-centers and moving dislocation may increase with their deformation.

(Received August 5, 2010; accepted September 15, 2010)

Keywords: Photoplastic effect, Plastic deformation, Alkali halides, Color centers

1. Introduction

Photoplastic effect (PPE) is a phenomenon where the flow stress of materials is appreciably affected by illumination of light with a wavelength around the intensive absorption. The phenomenon of photoplastic effect is reversible, that is, when the Light is turned off, the flow stress return to its expected initial value. Both positive (hardening) and negative (softening) photoplastic effects have been observed. The photoplastic effect first reported by Nadeau[1] has been extensively investigated in colored crystals of NaCl, KCl, KBr, and KI. The studies have explored important information on both the small

scale variation of the feature of the relevant defects and the chemical reaction of the color centers subjected to the optical excitations in the concerning various features, such as optical-, electrical- and mechanical-properties in the alkali metal colloid particles in densely colored alkali halides [2-11]. The plastic deformation of non metals has been the subject much research throughout the world for the last three decades. A wide variety of nonmetallic materials exhibit photoplastic behaviour, including alkali halides [NaF, KF, KI, NaCl, KBr (1), KCl [1,12-13]] , I - VII compounds [CuCl (14), CuBr [15]], II-VI semiconductors [ZnS (16), ZnO (16-18), ZnSe, ZnTe, Cdse, CdTe (16,18), CdS [16,18-20], HgCdTe[21]], III-V semiconductors GaAs(22), and group IV semiconductors Si [23]. The photoplastic effects in II-VI semiconductors have been reviewed by Takeuchi et al (24) and Ossipyan et al [25].

Because the photoplastic effect apparently deals with the interaction between charged dislocations and charged defects, it should have an important bearing on the plastic behaviour of dielectric materials. It should be especially relevant to irradiation or defect hardening as well as to the processes of annihilation and conversion of color centers induced by plastic deformation. Furthermore, it can also throw some light on the charge structure of dislocations in ionic compounds and provide a new tool for determining the charge. An ever-growing interest in investigating the PPE is also caused first by the need of a better understanding of the influence of the electronic states on the crystal plasticity and second by the fact that this effect underlies the degradation of opto-electronic devices.

The PPE in alkali halides containing F-centers has been attributed to the interaction between moving charged dislocations and defects, originating from photochemical reactions and bleaching of the F-centers due to optical excitation. Nadeau [1] has pointed out that there exist two photoplastic effects in alkali halides, i.e., a flow stress increases during light illumination (positive PPE) in twelve alkali halides such as KCI, KBr, NaBr, and so on, as well as a decrease of flow stress (negative PPE) in NaF, KF, etc. Since that time, much work has been done by various investigators [21, 26-30]. It has been considered that the positive PPE may give rise to retardation of the motion of the dislocations due to the F-center [1, 27,30], electron cloud around the dislocation [29] and the aggregation of the metallic colloids along the dislocation [31]. On the other hand, as to the negative PPE, Cabrera and Agullo-Lopez [29] have suggested that the interaction between an excess anion arrays along dislocations, which is caused by agglomeration of the excess action vacancies, and the anion vacancies resulting from the ionization of the F-centers may take place in the specimen doped with divalent captions. Korovkin and Soifer [32] have later concluded that their results are well explained by considering the interaction with the Z-type centers (complex of the F-center and divalent caption and caption vacancy [33].

In past significant experimental works have been made on the PPE in alkali halide crystals. It has been found that the photoplastic effect depends upon several parameters like strain level strain rated light intensity F-center concentration wavelength of the exciting light etc. to date, there is a few theories which explains satisfactorily the PPE in alkali halide crystals. Therefore it will be of worth to investigate theoretically the PPE in alkali halide crystals. The present paper reports the theory of positive photoplastic effect in colored alkali halide crystals and makes comparison between the theoretical and experimental results.

In previous investigation Hagihara and Okada [2] have proposed the theory of PPE which is based on the statistical dislocation dynamics whereby the motion of dislocations interacting with the optical sensitive defects is described by a master equation. Then by introducing several characteristic its interaction times, they have extended the theory to the PPE in alkali halides containing F-centres. We have proposed a new theory which begins with the interaction of dislocations with F-centers, hereby the population of dislocation band takes place. Subsequently by the exposure of deforming sample containing F-centers to light causes the creation of

F'-centers whereby the interaction between the negatively charged F'-centers and positively charged moving dislocations gives rise to the photoplastic effect. Although the interaction of dislocations with color centers is common, the mathematical approaches are quite different in both the theories.

2. Theory

From the several experiments, it has been confirmed that photoplastic effect (PPE) is closely related to the excitation of F-centers. Typical photochemical process of F-centers during F-bleach at around room temperature may be expressed as

$$2\mathrm{F} \xrightarrow{h\varpi} \mathrm{F}^1 + \mathrm{F}^+$$

Where F' is the center having two electrons in a negative ion vacancy and F^+ is the negative ion vacancy without trapped electrons.

Nadeau [1] has shown that F^+ center can be eliminated as hardening center at around room temperature. Thus, the interaction between the positively charged dislocations and negatively charged F'- centers may be responsible for the PPE.

On illuminating F-centers, equilibrium state of F === F' is accomplished in a few seconds. Dislocations are assumed to have constant equilibrium charges (ρ) per unit length and they can interact with F'-centers only in a cylindrical region of radius R around dislocations with appropriate mean strength ($\bar{f}$). Dislocation densities are expected to increase with the increase of the plastic strain (ε). In this case, the interaction region could be expected to increase in the crystal with plastic deformation.

Ermakov et. al. [27] has shown that the photo-obstacle centers, responsible for PPE in alkali halide crystals are F-centers.

In PPE experiment, the color centers are bleached due to deformation as well as due to the F-light illumination. Now, we will estimate these bleaching, one by one.

When a colored alkali halides crystal is plastically deformed, the movement of dislocations takes place. The moving dislocations may captures electrons from the colour centers and may subsequently transport the captured electrons to hole centers, deep traps and other compatible traps in the crystal [34]. As a matter of the fact, deformation bleaching of the coloration in alkali halide crystals may take place. Suppose a crystal contains N_d dislocations of unit length per unit volume. When N_d dislocations will move through a distance dx, then the area traps out by the dislocations will be $N_d dx$. The deformation bleaching in colored alkali halide crystals takes place due to the transfer of electrons from F-centers to the dislocation band and their subsequent recombination with other centers. Near an edge dislocation, some of the F-centers lie in the expansion region and some of the F-centers lie in the compression region. In the expansion region, the energy gap between ground state of F-centres level and dislocation band which lies just above the ground state of F-center level [35-36] deceases due to the decrease in local density of the crystal, however, the energy gap between the ground state of F-centre level and dislocation band increases in the compression region of the dislocation due to the increase in the local density of the crystal. Since, there is a greater probability of the transfer of electrons from the F-centers lying in the expansion region rather than from the compression region of the edge dislocations, the interaction volume may be taken only along the expansion region of the crystals. Thus, the volume in which N_d dislocations interact while moving through a distance dx may be given by

$N_d dx r_F$, where r_F is the distance upto which a dislocation can interact with the color centers.

If n_F is the number of F- centers in unit volume, then the number of color centers interacting with the dislocations will be $N_d n_F r_F$ dx. If a dislocation moves the distance dx in time dt, then the rate of interaction g_i of the number of F-centers with dislocations may be given by

$$g_i = N_d n_F r_F \frac{dx}{dt} = N_d n_F r_F v_d$$

or

$$g_i = \frac{\dot{\varepsilon}}{b} n_F r_F \tag{1}$$

Where v_d is the average velocity of dislocations, $\dot{\varepsilon} = N_d b v_d$ is the strain rate and b is the Burgers vector.

Now, there are two possibilities: Firstly, the electron from interacting F-center may get transferred to the dislocation band laying just above the F-center level, secondly, the electron from the F-centre may return to the negative ion vacancy. If α_1 is the probability of the transfer of F-centre electrons to the dislocation band during the interaction of dislocations with F-centers electrons to the dislocation band during the interaction of dislocations with F-centers and α_2 is the back to the negative ion vacancies, then we may write the following rate equation

$$\frac{dn_i}{dt} = g_i - (\alpha_1 + \alpha_2)n_i = g_i - \frac{n_i}{\tau i} \qquad (2)$$

Where $1/(\alpha_1 + \alpha_2) = \tau_i$ is the lifetime of the interacting F-center, and n_i is the number of F-centers interacting with dislocations at any time t.

For $n_i = 0$ at t = 0, the integration of above equation gives

$$n_i = \frac{g_i}{(\alpha_1 + \alpha_2)}\left[1 - \exp[-(\alpha_1 + \alpha_2)]t\right] \qquad (3)$$

From equations (2) and (3), the rate of generation of electrons in the

$$g = \frac{a_1 g_i}{(\alpha_1 + \alpha_2)}\left[1 - \exp[-(\alpha_1 + \alpha_2)t\right] \qquad (4)$$

For $(\alpha_1 + \alpha_2)t >> 1$, i.e., in equilibrium the rate of generation of electrons in the dislocation band may be given by

$$g = \frac{\alpha_1 g_i}{(\alpha_1 + \alpha_2)}$$

or

$$g = \frac{\alpha_1}{(\alpha_1 + \alpha_2)}\frac{\dot{\varepsilon}}{b} r_F n_F \qquad (5)$$

or

$$g = \frac{\dot{\varepsilon}}{b} p_F r_F n_F$$

Where $p_F = \frac{\alpha_1}{(\alpha_1 + \alpha_2)}$, is the dislocation capture probability of F-centres electrons.

It has been found that only few electrons near the core of moving edge dislocations are bleached during the deformation of crystals [34, 37]. Thus, it seems that when $\alpha_1 << \alpha_2$, p_F may be written as

$$p_F = \frac{\alpha_1}{\alpha_2}, \qquad (6)$$

Since there is a band gap of E_a between the bottom of dislocation band and the ground state energy level of the interacting F- entres lying in the expansion region of an edge dislocation [34], the activation process should be involved in transferring the electrons from interacting F-centres to the dislocation band. Thus, α_1 should be temperature dependence and it may be given by

$$\alpha_1 = \alpha_1^0 \exp(-E_a/kT) \qquad (7)$$

where α_1^0 is a constant.

From equations (6) and (7), P_F may be expressed as

$$p_F = p_F^0 \exp(-E_a/kT) \qquad (8)$$

Where $p_F^0 = \frac{\alpha_1^0}{\alpha_2}$.

When the moving dislocations containing electrons encounter defect-centres like hole centres, deep traps and other compatible traps, the electrons are captured by these centres. As such the rate equation may be written as

$$\frac{dn_d}{dt} = g - \sigma_1 N_1 v_d n_d - \sigma_2 N_2 v_d n_d - \sigma_3 N_3 v_d n_d = g - \frac{n_d}{\tau_d} \qquad (9)$$

Where

$$\tau_d = \frac{1}{(\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3)v_d} \qquad (10)$$

is the lifetime of electrons in the dislocation band, n_d is the number of electrons in the dislocation band at any time t, σ_1, σ_2 and σ_3 are the cross-sections and N_1, N_2 and N_3 are the densities of hole centres, deep traps and other compatible traps, respectively. The drift velocity of electrons has been taken as the velocity of dislocations because the dislocation captured electrons are moving with the dislocations. Here, the compatible trap means the traps whose electron-capture probability greater than that of the dislocations. It should be noted that the vacant negative ion vacancies have nearly the same probability of electron-capturing and electron-detrapping hence; their presence may not affect significantly the recombination processes [38].

Since τ_d is shorter as compared to the time needed for generating the electrons in the dislocation band [34] equilibrium will be established quickly between the rate generation and rate of recombination of dislocation electrons, and thus from eq. (9), we get

$$n_d = g\tau_d \qquad (11)$$

From eqs. (4) and (11), we get

$$n_d = \frac{p_F r_F n_F \dot{\varepsilon} \tau_d}{b}\left[1 - \exp(-t/\tau_i)\right] \qquad (12)$$

Now, the rate of recombination of dislocation electro with defect centres may be given by

$$R_d = \frac{n_d}{\tau_d} = \frac{p_F n_F r_F \dot{\varepsilon}}{b}[1 - \exp(-t/\tau_i)] \qquad (13)$$

As a crystal of volume V will contain $N_d V$ dislocation the value of R_d may be given by

$$R = R_d V = \frac{p_F n_F r_F \dot{\varepsilon} V}{b}[1 - \exp(-t/\tau_i)] \qquad (14)$$

Equation (14) shows that for a given strain rate, R will initially increase with time and then it will attain saturation value for longer duration of the deformation time The saturation value of R may be expressed as

$$R_S = \frac{p_F n_F r_F \dot{\varepsilon} V}{b} \tag{15}$$

The above equation shows that in equilibrium, the rate of recombination is equal to the rate of generation electrons in the dislocation band.

The number of dislocation electrons recombined with the defect centres will be equal to the number of F-centres bleached. Thus, the number of F-centres bleached by deformation for the change of strain from ε to (ε + Δε) of the crystal may be given by

$$-\Delta n_{FD} = \frac{p_F n_F r_F}{b} \int_{\varepsilon}^{\varepsilon+\Delta\varepsilon} \frac{d\varepsilon}{dt} dt$$

or

$$\Delta n_{FD} = -\frac{p_F n_F r_F}{b} = -D n_F \Delta\varepsilon \tag{16}$$

where $D=(p_F r_F)/b$, is the coefficient of deformation bleaching.

It is known that absorption coefficient of coloured alkali halide crystals is directly proportional to the density of colour centres [38]. If A is the cross-sectional area of the F-centre for the optical excitation, then the absorption coefficient will be $A_n F$. For the intensity I_L of the incident radiation on the sample, the rate of generation of excited F-centres by photo-ionization may be written as

$$g_e = A n_F I_L \tag{17}$$

If B is the probability of ionization of excited F-centres, then the rate of optical bleaching of F-centres is given by

$$g_{op} = BA n_F I_L \tag{18}$$

In PPE experiment, the sample is not exposed to light continuously, however, it is exposed frequently for a short interval of time. If f is the ratio of the total time duration to which the sample is exposed to light and the total duration of deformation, then the number of F-centres bleached optically for the change of strain from ε to (ε + Δε) of the crystal may be given by

$$-\Delta n_{FOP} = fBA n_F I_L dt$$

or

$$-\Delta n_{FOP} = \frac{fBA n_F I_L \Delta\varepsilon}{\dot{\varepsilon}} = \xi n_F \Delta\varepsilon \tag{19}$$

where $\xi = \frac{fABI_L}{\dot{\varepsilon}}$.

From eqs. (16) and (19), the number of F-centres bleached due to the deformation and F-light illumination is given by

$$-\Delta n_F = -(\Delta n_{FD} + \Delta n_{FOP}) = \alpha n_F \Delta\varepsilon \tag{20}$$

where $\alpha=(D+\xi)$.

Integrating eq. (20) and taking $n_F = n_{FO}$, at ε = 0, we get

$$n_F = n_{FO} \exp(-\alpha\varepsilon) \tag{21}$$

where n_{FO} is the density of F-centres in the undeformed crystal.

In the PPE experiment, the sample is excited by exposing it to light at $t = t_c$.

If γ_1 is the rate constant for the transformation of excited F-centers into normal F-centers, and γ_2 is the rate constant for the formation of F'-centres from the excited F-centres, then we may write the following rate equation

$$\frac{dn_e}{dt} = g_e - \gamma_1 n_e - \gamma_2 n_e \tag{22}$$

where n_e is the number of excited F-centers at any time t.

As the lifetime of F'-centre electrons is less than 1 sec at RT, one may think that there may also be some contribution of F'- centres in giving rise to excited F-centres. However, such contribution by F'-electrons may be neglected because of the following facts (i) in PPE experiments the sample is mostly exposed to F-light whereby the light absorption by F'-centres is not significant, (ii) the number of F-centers is significantly higher than the number of F'-centers for all the time values after exposure of the sample to F-light, and (iii) the excitation of F-centers generated during exposure of the sample to F-light, will give excited F'-centers and not the excited F-centers. It is to be noted that the formation of F'-centers during exposure of the sample to light plays an important role in the phenomenon of PPE. This fact has been discussed in the following paragraphs.

In the PPE measurement the sample is deformed at a very slow strain rate. Therefore, we may assume that for the time duration during which a single PPE measurement is made, the strain ε may be assumed to be constant. Thus, integrating eq. (22) and taking n = 0, at $t = t_c$, we get

$$n_e = \frac{g_e}{(\gamma_1+\gamma_2)} \left\{1-\exp[-\gamma_1+\gamma 2)(t-t_c)\right\} \tag{23}$$

Thus, the rate of generation of F'-centres may be written as

$$G_{F'} = \gamma_2 n_e = \frac{\gamma_2 g_e}{(\gamma_1+\gamma_2)} \left\{1-\exp[-(\gamma_1+\gamma_2)(t-t_c)]\right\} \tag{24}$$

As $1/(\gamma_1+\gamma_2)$, the lifetime of excited F-centres is very short (less than a microsecond), $(\gamma_1+\gamma_2)$ is very high and in equilibrium, the rate of generation of F'-centers may be written as

$$G_{F'S} = \frac{\gamma_2 g_e}{(\gamma_1+\gamma_2)} \tag{25}$$

If $\tau=1/\beta$ is the lifetime of normal F'-centres, then we can write

$$\frac{dn_{F'}}{dt} = G_{F's} - \beta n_{F'} \tag{26}$$

where $n_{F'}$ is the number of F'-centres at any time t. Since the $n_{F'}$ will depend recombination of on the rate of generation and rate of F'-centres, the contribution of F-centre interacting with dislocations is not taken into account to the right hand side of eq. (26).

Integrating eq. (26) and taking $n_{F'}$=O at t=$t_{c'}$ the time at which light is turned on, we get

$$n_{F'}=\frac{G_{F's}}{\beta}\left\{1-\exp[-\beta(t-t_c)]\right\} \quad (27)$$

For longer duration of exposure to light, i.e., in equilibrium, eq. (27) may be expressed as

$$n_{F's}=\frac{G_{F's}}{\beta} \quad (28)$$

If r_i is the radius of interaction of dislocations with the F' –centres produced during exposure of the sample to light, then similarly to that of the F-centres, the rate of generation of interacting F'-centres may be expressed as

$$g_1^{'}=\frac{\dot{\varepsilon}}{b}n_{F'S}r_i$$

or

$$g_1^{'}=\frac{\dot{\varepsilon}}{b}\frac{G_{F'S}r_i}{\beta}$$

or

$$g_1^{'}=\frac{\dot{\varepsilon}}{b}\frac{r_i}{\beta}\frac{\gamma_2 g_e}{(\gamma_1+\gamma_2)}$$

or

$$g_1^{'}=\frac{\dot{\varepsilon}r_i\gamma_2 An_{FO}I_L}{b\beta(\gamma_1+\gamma_2)}\exp(-\alpha\varepsilon) \quad (29)$$

It is to be noted that the electrons of interacting F'-centres lie in the normal energy level of F'-centres. However, some of the interacting F'-centres may be captured by dislocations and the electrons of such dislocation-captured F'-centres may lie in the traps slightly different from normal F'-level [39]. Since the F'-centres captured by dislocations may only pin the dislocations, the PPE will depend only on the number of

F'-centres captured by the dislocations [12,40].

If δ_1 is the rate constant for the transformation of interacting F'-centres into F-centres by their recombination F^+-centres ($F'+F^+ \rightarrow 2F$) and δ_2 is the rate constant for the capture of F'-centres by the dislocations, then we can write the following equation

$$\frac{dn_1^{'}}{dt}=g_1^{'}-\delta_1 n_1^{'}-\delta_2 n_1^{'}$$

or

$$\frac{dn_1^{'}}{dt}=g_1^{'}-\delta_1 n_1^{'}$$

where $\delta=(\delta_1+\delta_2)$.

Now, substituting the value of g! from eqn. (29), we get

$$\frac{dn_1^{'}}{dt}=\frac{\dot{\varepsilon}r_i\gamma_2 An_{FO}I_L}{b\beta(\gamma_1+\gamma_2)}\exp(-\alpha\varepsilon)=\delta n_1^{'} \quad (30)$$

where $n_1^{'}$ is the number of interacting pi-centres at any time t and 1/δ is the lifetime of interacting F'-centres or the pinning time of dislocations [41].

In certain crystals, the radius r_i of interaction between the charged dislocations and pi-centres, may increase significantly with the deformation of crystals. This may due to the increase in linear charge density of moving dislocations with deformation of crystals [42], as well as due to the increasing concentration of vacancies with the deformation of crystals. If 0: is the deformation coefficient of the increase in radius of interaction between the dislocations and F'-centres, we may write

$$r_i=r_i^0(1+\alpha_0\varepsilon) \quad (31)$$

where r_0^0 is the value of r_i for undeformed crystals.

For $\alpha_0\varepsilon<<1$, eg. (31) may be expressed as

$$r_i = r_i^0\exp(\alpha_0\varepsilon) \quad (32)$$

From eqs. (30) and (32), we get

$$\frac{dn_1^{'}}{dt}=\frac{\dot{\varepsilon}r_i\gamma_2 An_{FO}I_L}{b\beta(\gamma_1+\gamma_2)}\exp(-\alpha\dot{\varepsilon}t)-\delta n_1^{'} \quad (33)$$

where $\alpha_e=(\alpha-\alpha_0)$.

Integrating eq. (33) and taking $n_1^{'}$ = 0 at t = 0, we get

$$n_i^0 = \frac{\dot{\varepsilon}r_i\gamma_2 An_{FO}I_L}{b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e\dot{\varepsilon})}[\exp(-\alpha_e\dot{\varepsilon}t)-\exp(-\delta t)] \quad (34)$$

Thus, the rate of generation of (34) dislocation-capture F'-centres may be written as

$$g_d = \delta_2 n_i^{'} = \frac{\delta_2\dot{\varepsilon}r_i\gamma_2 An_{FO}I_L}{b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e\dot{\varepsilon})}[\exp(-\alpha_e\dot{\varepsilon}t)-\exp(-\delta t)]$$

or

$$g_d == \frac{\delta_2\dot{\varepsilon}r_i\gamma_2 An_{FO}I_L}{b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e\dot{\varepsilon})}[\exp(-\alpha_e\dot{\varepsilon}t)-\exp(-\frac{\delta\varepsilon}{\dot{\varepsilon}})] \quad (35)$$

If $\tau_d^{'}=1/\delta_0$ is the life time of dislocation captured F'- centres, then we can write the following rate equation

$$\frac{dn_d}{dt} = g_d - \delta_0 n_d \quad (36)$$

where n_d is the dislocation captured F-centres at any time t.

For low strain-rate, g_d may be taken as constant during the measurement of single PPE, which is completed in a short duration. Thus, integrating eq. (36) and taking n_d = 0, at t ='c' we get

$$n_d = \frac{d_d}{\delta_0}\left\{1-\exp[-\delta_0(t-t_c)]\right\} \quad (37)$$

From eqs. (35) and (37), we get

$$n_d = \frac{\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon})} [\exp(-\alpha_e \dot{\varepsilon}) - \exp(-\frac{\delta\varepsilon}{\dot{\varepsilon}})]$$

$$\{1-\exp[-\delta_0 (t-t_c)]\} \tag{38}$$

Total Coulomb force (W) between positive charge p on a dislocation and negative charge surrounding the dislocations (the F'-centres in the present case) may be written as

$$W = c\rho e \sum_{j=1}^{n_d} \frac{1}{r_j^2} \tag{39}$$

where n_d is the number of dislocation captured F'-centres in unit volume of the specimen, r. is the distance between a dislocation and an F'-centre and C is a constant. It is to be noted that the number of F-centre electrons trapped by dislocations may be smaller than the negative ion vacancies giving rise to positive charges to dislocations, so that dislocation charges might not be influenced by the F-centre electrons trapped by the dislocations [30].

The radius of interaction r_i in which F'-centre can interact with dislocation is assumed to be obtained from

$$W_0 = \frac{C\rho e n_d}{r_i^m} = \overline{f} n_d \, (m = 1-2) \tag{40}$$

where $\overline{f}$ is an appropriate mean strength between a dislocation and an F'-centre and it is given by

$$\overline{f} = C\rho \frac{e}{r_i^m} \tag{41}$$

It is known that the force caused by a uniform shear stress σ is F = bσ per unit length of dislocation [43]. Thus, the photoplastic effect Δσ may be given by

$$\Delta\sigma = (\pi r_1^2 .1)\frac{W_0}{b} = \frac{\pi r_i^2 \overline{f} n_d}{b} = D n_d \tag{42}$$

where $D = (\pi r_i^2 \overline{f})/b$, is a constant.

Now, substituting the value of nd from equation (38) in eq. (42), we get

$$\Delta\sigma = \frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} [\exp(-\alpha_e \dot{\varepsilon}) - \exp(-\frac{\delta\varepsilon}{\dot{\varepsilon}})]$$

$$\{1-\exp[-\delta_0 (t-t_c)]\} \tag{43}$$

or

$$\Delta\sigma = (\Delta\sigma)_s \{1-\exp[-\delta_0 (t-t-_c)]\} \tag{44}$$

Where

$$(\Delta\sigma)_s = \frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} [\exp(-\alpha_e \dot{\varepsilon}) - \exp(-\frac{\delta\varepsilon}{\dot{\varepsilon}})] \tag{45}$$

It is to be noted that in the measurement of PPE, the sample is deformed at a very low strain rate and the sample is exposed to light for a short duration. Thus, for the time duration during which a single PPE measurement is made, the strain £; may be taken to be constant and thereby $(\Delta\sigma)_s$ may be treated as constant.

Rise of PPE

For smaller values of ($t-t_c$), eq. (44) gives

$$\Delta\sigma = (\Delta\sigma)_s \delta_0 (t-t_c) \tag{46}$$

Equation (46) indicates that when light will be turned on, initially, $\Delta\sigma$ should increase linearly with ($t-t_c$).

It is evident from eq. (44) that for longer duration time $\delta_0 t$ will be greater than 1 and $\Delta\sigma$ will attain a saturation value $(\Delta\sigma)_s$, given by eq. (45).

2.1 Decay of Photoplastic effect

When the light source will be turned off at t = t', then $I_L = 0$ and therefore $g_d = 0$, and from eq. (36), we get

$$\frac{dn_d}{dt} = -\delta_0 n_d \tag{47}$$

Integrating eq. (47) and taking $n_d = n_{d0}$ at $t = t_c'$ we get

$$n_d = n_{do} \exp[-\delta_0 (t-t_c')) \tag{48}$$

where n_{do} is the saturation value of n_d, i.e, the value of n_d at $\delta_0 t >> 1$, and from eq. (38), it can be expressed as

$$n_{do} = \frac{\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} [\exp(-\alpha_e \dot{\varepsilon}) - \exp(-\frac{\delta\varepsilon}{\dot{\varepsilon}})] \tag{49}$$

Thus, Δσ may be expressed as

$$\Delta\sigma = D n_D = D n_{do} \exp[-\delta_0 (t-t_c')] \tag{50}$$

or

$$\Delta\sigma = \frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} [\exp(-\alpha_e \dot{\varepsilon}) - \exp(-\frac{\delta\varepsilon}{\dot{\varepsilon}})] \exp[-\delta(t-t_c')] \tag{51}$$

or $\Delta\sigma = (\Delta\sigma)_s \exp[-\delta_0 (t-t_c')]$

The above equation shows the exponential decay of Δσ, where the decay time will be controlled by the lifetime of the dislocation-captured F' -centres.

2.2 Strain dependence of PPE

It is evident from eq. (45) that $(\Delta\sigma)_s$ is zero for ε = 0 as well as for ε = ∞. Thus $(\Delta\sigma)_s$ should be maximum for a particular value of $\varepsilon = \varepsilon_m$.

As $(\Delta\sigma)_s$ will be maximum, when $\frac{d(\Delta\sigma)_s}{d\varepsilon} = 0$, eq. (45) gives

$$\alpha_e \exp(-\alpha_e \varepsilon_m) = \frac{\delta}{\dot{\varepsilon}} \exp\left(-\frac{\delta\varepsilon_m}{\dot{\varepsilon}}\right) \tag{53}$$

or

$$\varepsilon_m = \frac{1}{\left(\frac{\delta}{\varepsilon} - \alpha_e\right)} \ln\left[\frac{(\delta/\dot{\varepsilon})}{\alpha_e}\right] \tag{54}$$

Thus, form eqs. (4.53) and (4.54), the maximum value of $(\Delta\sigma)_s$ may be expressed as

$$(\Delta\sigma)_{max} = \frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} \left[\frac{\delta}{\alpha_e \dot{\varepsilon}} \exp\left(\frac{-\delta\varepsilon_m}{\dot{\varepsilon}}\right) - \exp\left(\frac{-\delta\varepsilon_m}{\dot{\varepsilon}}\right)\right]$$

$$\frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} \left\{\left(\frac{\delta}{\alpha_e \dot{\varepsilon}} - 1\right) \exp\left[\frac{\delta}{\dot{\varepsilon}\left(\frac{\delta}{\dot{\varepsilon}} - \alpha_e\right)} \ln\left(\frac{-\delta/\dot{\varepsilon}}{\alpha_e}\right)\right]\right\}$$

For $\delta/t >> \alpha_e$, we have

$$(\Delta\sigma)_{max} = \frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} \left[\frac{\delta-\alpha_e \dot{\varepsilon}}{\alpha_e \dot{\varepsilon}}\right] \frac{\alpha_e \dot{\varepsilon}}{\delta}$$

or

$$(\Delta\sigma)_{max} = \frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} \tag{55}$$

For low values of ε, $\alpha_e \varepsilon << 1$, and eq. (45) gives

$$(\Delta\sigma)_s = \frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} \left[1 - \exp\left(\frac{\delta\varepsilon}{\dot{\varepsilon}}\right)\right] \tag{56}$$

Equation (56) indicates that for low strain $(\Delta\sigma)_s$ should initially increase linearly with strain and then it should attain a saturation value.

For high value of ε, $\frac{\delta\varepsilon}{\dot{\varepsilon}} >> 1$, and eq. (4.45) may be expressed as

$$(\Delta\sigma)_s = \frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} \exp(-\alpha_e \varepsilon) \tag{57}$$

It is evident from eq. (57) that for high values of strain $(\Delta\sigma)_s$ should decrease exponentially with strain, where the decay constant will be controlled α_e.

3. Experimental support to the proposed theory

3.1 Temporal caracteristics

Fig. 1 shows the PPE for NaCl and KCl crystals measurec by Inoue et al [30] at room temperature and Fig. 2 shows the PPE of KCl crystals measured by Hagihara [40] at low temperatures. When the samples are illuminated during deformation, rise in flow stress is observed. A region of steady flow state keeps during illumination. When the light is turned off, the stress decreases exponentially to the initial stress level. The results illustrated in Fig. 2 indicate the linear increase of $\Delta\sigma$, when the light is turned on. Such linear increase of $\Delta\sigma$, attainment of saturation value and exponential decay indicated by equations (46), (44) and (52), respectively. The decay time $\tau_d' = 1/\delta_0$ of dislocation captured F'centres, is determined from the decay time of PPE and it is found to be 2.8 sec and 2.2 sec for 166K and 233K, respectively. The decay time of dislocation captured F'-centres determined from the rise PPE in KCl crystals with time is determined using eq. (44) and is found to be 2.5 sec and 2.0 sec for 166K and 233K respectively. The decay time of dislocation-captured F'-center is determined for NaCl crystals using eq. (52) and the result reported by Nadeau [1] and it is found to be 1.6 sec. Korovkin [44] has reported that the decay time of PPE is of the order 10 sec for NaCl crystals.

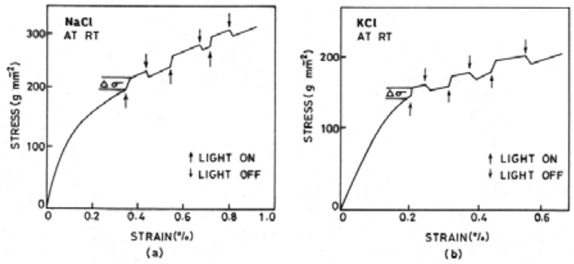

Fig. 1. Photoplastic effect for (a) NaCl and (b) KCl measured at room temperature. Turn on and off of llumination are marked by arrows (strain rate 4 –1 1.2 X 1s) [30].

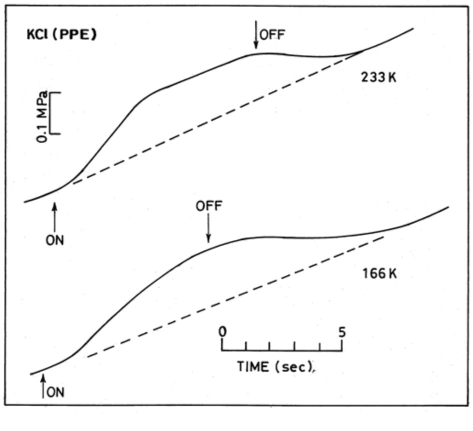

Fig. 2. Typical examples of the PPE in coloured KCl 5 –1 strain rate 5 X 10 s) [40].

3.2 Deformation characteristics

Figs. 3 and 4 show the dependence of Δσ on the strain level, for KCl and NaCl crystals, where the experimental data have been taken from the results reported by Inoue et al [30 Inoue et. al. 1979] and the theoretical data are based on equation (45), here $\Delta\sigma_0$ corresponds to the value of $\Delta\sigma$ for zero strain level. Figs. 3 shows that for KCl crystals, $\Delta\sigma/\Delta\sigma_0$ is optimum for a particular strain and Figs. 4 shows that for NaCl crystals, $\Delta\sigma/\Delta\sigma_o$ initially increases at a fast rate and then it increases at a slow rate.

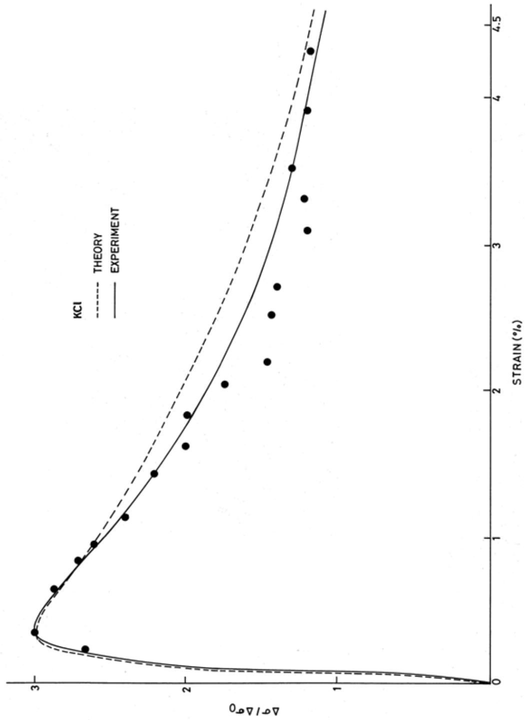

Fig. 3. The dependence of the photoplastic effect on strain level for KCl crystals [30].

Figs. 3 and 4 show a good agreement between the theoretical and experimental results. The value of α_e is found to be 22.75 for KCl crystals and –1.87 for NaCl crystals. This fact indicates that r_i^0 increases significantly with the deformation of NaCl crystals, so that the value of α_e becomes negative. On the other hand, the change of r_i^0 with deformation is less for KCl crystals, so that α_e is positive. Practically the value of α_e may be found higher due to the optical bleaching occurring during the PPE measurements.

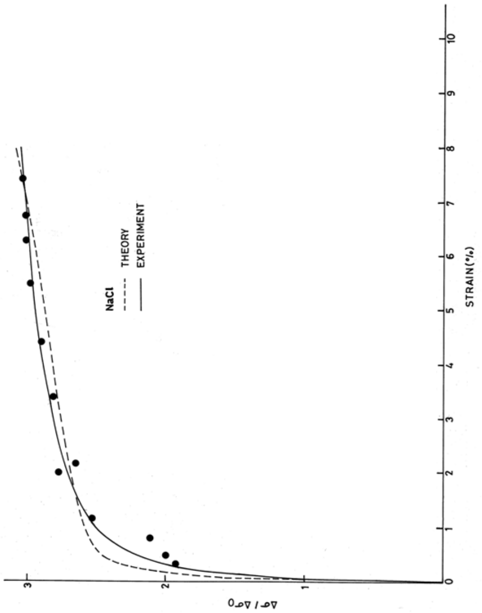

Fig. 4. The dependence of the photoplastic effect on strain level for NaCl crystal [30].

Using the curve-fitting technique in Figs. 3 and 4, the value of $\delta/\dot{\varepsilon}$ is found to be 1350 and 2000 for KCl and NaCl crystals, respectively. For $\dot{\varepsilon} = 1.2x10^{-4}s^{-1}, \delta$ comes out to be 0.16 and 0.04 s^{-1} for KCl and NaCl crystals, respectively. This gives that the lifetime of interacting F-centres or pinning time of dislocations is 6.25 and 4.16 sec, for KCl and NaCl crystals, respectively. This is comparable with the experimental results [42]. It is to be noted that the pinning time of dislocations depends on temperature, strain-rate and density of colour centres in the crystals.

3.3 Strain-rate characteristics

Fig. 5 shows the strain-rate dependence of PPE for coloured KCl crystals where the experiment was carried out by Hagihara et al [40] at 166 K. It is seen from the figure that increases linearly with the strain rate. For a strain rate $\dot{\varepsilon} = 1.2x10^{-4}s^{-1}$ the values of 0 are 0.16 and 0.04 s for KCl and NaCl crystals, respectively. The values of determined using the results of Figs. 3 and 4 and expression (45) are found to be 22.75 and -1.87 for KCl and NaCl crystals, respectively. These data clearly indicates that δ » $\alpha_e \dot{\varepsilon}$ and thus eq. (45) supports the linear increase of $\Delta\sigma$ with the strain rate.

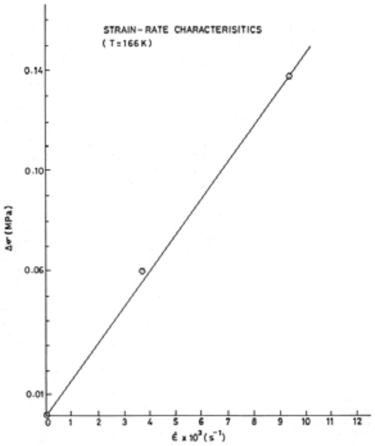

Fig. 5. Dependence of PPE in KCl crystals on the strain rate.

3.4 Spectral characteristics

Figs. 6 (a) and (b) show the dependence of the magnitude of $\Delta\sigma/\Delta\sigma_F$ on the wavelength (or photon energy) of exciting light for NaCl and KCl crystals, respectively, where $\Delta\sigma_F$ is the magnitude for $\Delta\sigma$ given at the same strain level in the case of F-light illumination, 465 nm for NaCl and 565 nm for KCl, respectively. Solid curve indicates the optical absorption spectra near the F-band peak. For NaCl [Fig. 6 (a)], a shape of $\Delta\sigma_F$ is close to the F-band in the short wavelength side but in the long wavelength side, $\Delta\sigma/\Delta\sigma_F$ does not fit to F-band. On the other hand, for KCl [Fig. 6(b)], $\Delta\sigma/\Delta\sigma_F$ has a similar shape of the F-band in the region of long wavelength, and has a different shape from the F-band in the short wavelength side. However, for both crystals, the wavelength in which the maximum value of $\Delta\sigma/\Delta\sigma_F$ appears coincide to the F-band peak. Both curves roughly resemble the respective F-absorption bands. Such fact is expected from the proposed theory.

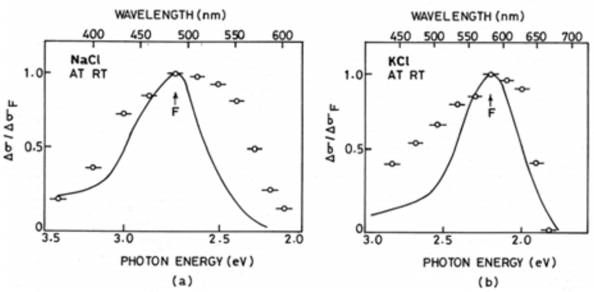

Fig. 6. Dependence of the photoplastic effect on wavelength (or photon energy) of exciting light for (a) NaCl and (b) KCl crystals. F-centre concentrations (N_F) for NaCl and KCl are 1.0 x10 $^{1\ 7}$cm^{-3} and 4.7×10^{16} cm^{-3}, respectively. Solid curves correspond to the optical absorption spectra near the F-band peak [30].

3.5 Light intensity characteristics

Fig. 7 (a) and (b) shows the dependence of a photoplastic effect on light intensity for KCl and KI crystals. It is seen that, initially, the PPE increases linearly with the light intensity and then it tends to attain a saturation value for higher intensity of the light. It seems that for high light intensity, a large numbers of F' -centres are formed, whose recombination with F$^+$-centres may cause the occurrence of saturation in $\Delta\sigma$ when the rate of generation of F' -centres will become equal to their rate of recombination initially, the recombination is negligible and an increase in with the light intensity is observed. It is evident from eq. (45) that for a given strain, the PPE should increase linearly with the light intensity IL.

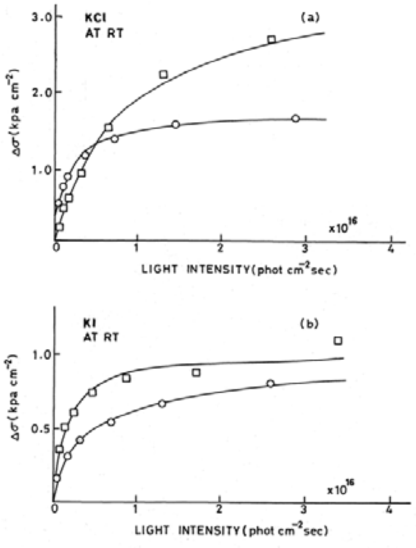

Fig. 7. Dependence of the photoplastic effect on light intensity for (a) KCI and (b) KI crystals. For each crystal, F-centre concentrations and wavelengths of excitation light are the following KCl : $n_F = 1.4$ x 10^{16} cm^{-3}, λ = 555 m μ (O); $n_{F_{17}}$ $3x10^{17}$ cm^{-3}, *, λ = 435 mμ (o). KI :* $n_{F_{-3}}$ $=2x10^{17}$ cm^{-3}*, λ = 435 mμ(o);*

n_F $=2x10^{17}$ cm^{-3}*, λ = 570 mμ(o) [29].*

3.6 Coloration characteristics

Fig. 8 shows the dependence of the magnitude Δσ a of the photoplastic effect on F-centre concentration for o-irradiated KI crystal. Other alkali halide crystals show the similar behaviour. The concentration is varied between 5 X 105 and 3 X 1017 cm^{-3} the main results are the following:

(i) For wavelengths aside from the F-band peak, monotonic rise in Δσ with concentration is observed. Although the rate of increase in Δσ diminishes at high F-centre concentrations, a definite saturation has not been found.

(ii) For light of wavelength corresponding to the F-band peak, the effect increases rapidly with concentration upto a maximum and then decreases for higher concentrations.

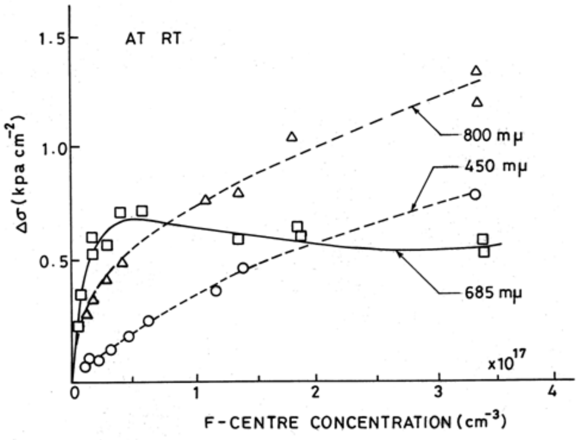

Fig. 8. Dependence of the photoplastic effect on F-centre concentration for KI. Wavelength of excitation light is marked on each curve [29].

It is to be noted that the maximum of $\Delta\sigma$ occurs for an F-centre concentration which corresponds to an optical density - 1.5. The same conclusion is valid for all alkali halides used in the experiments. This fact suggests that the maximum of $\Delta\sigma$ as well as the subsequent decay should be attributed to non-uniform illumination of the samples. It is clear that for optical densities most light is absorbed in a small fraction of the sample thickness. Equation (45) indicates that for a given strain the PPE should increase lineary with the initial colour centre concentration n_{FO}. This is practically true for these samples having low concentration of colour centres where the effect of non-uniform coloration does not occur.

3.7 Dimension characteristics

Confirmation of the idea described above in Section 3.6, can be obtained by studying the dependence of the photoplastic effect on sample thickness while keeping the F-centre concentration constant. Experimental data for KCl are shown in Fig. 9. One should then conclude that meaningful information on the role of the F-centre concentration can only be obtained if one works with low optical densities to void the above "geometry" effect. In this regard, the curves in Fig.8 taken with alight of wavelength aside from the F-peak (optical density < 0.5) are only relevant to that purpose. It is clear that the photoplastic effect increases monotonically with F-centre concentrations.

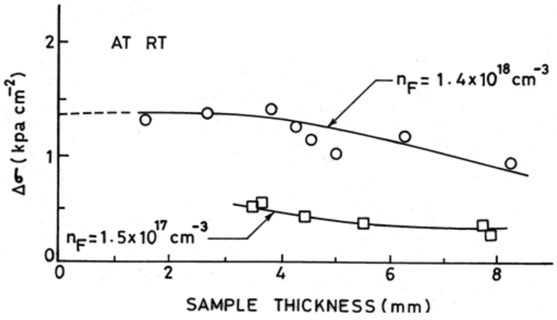

Fig. 9. Dependence of the photoplastic effect in KCI on sample thickness (in the direction of light illumination). Other dimensions are kept constant [29].

3.8 Thermal characteristics

The dependence of the magnitude of the photoplastic effect on temperature is shown in Fig. 10 (a, b) for KBr and KI crystals. Monochromatic light has been used for all experiments. It is seen $\Delta\sigma$ is maximum for a particular temperature of the crystals. It is evident form eq. (45) that the temperature dependence $(\Delta\sigma)_s$ may primarily be due to the temperature dependence of $n_{FO}, r_i^o, \gamma_2/(\gamma_1+\gamma_2), \delta_2, \delta$ and δ_0. As the value of n_{FO} decreases with temperature, however, the value of $r_i^0, \gamma_2/(\gamma_1+\gamma_2), \delta_2 \delta$ and δ_0 increase with temperature, $(\Delta\sigma)_s$ should be optimum for a particular temperature of the crystal.

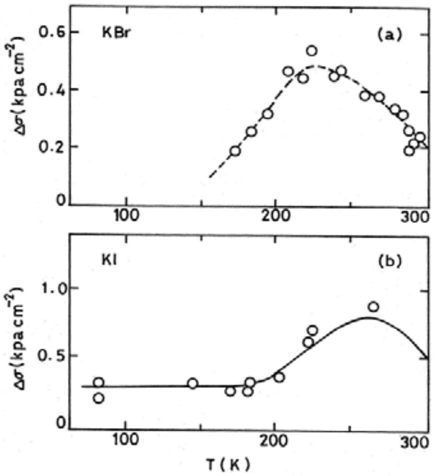

Fig. 10. Dependence of the photoplastic effect on temperature. F-center concentration and light wavelengths are the following: KBr : $n_F = 2\times10^{17}\,\mathrm{cm}^{-3}$, *$\lambda$ = 580 mμ(o); KI :* $n_F = 2\times10^{17}\,\mathrm{cm}^{-3}$, *$\lambda$ = 585 mu [29].*

Fig. 11 shows the kinetics of the stress decay after light Fig.12 is turned off for additively coloured KCI crystals shows that the temperature dependence of the rate constant for stress decay follows the Arrhenius plot. This fact shows that the lifetime of dislocation-captured F’-centres decreases with increasing temperature of the crystals.

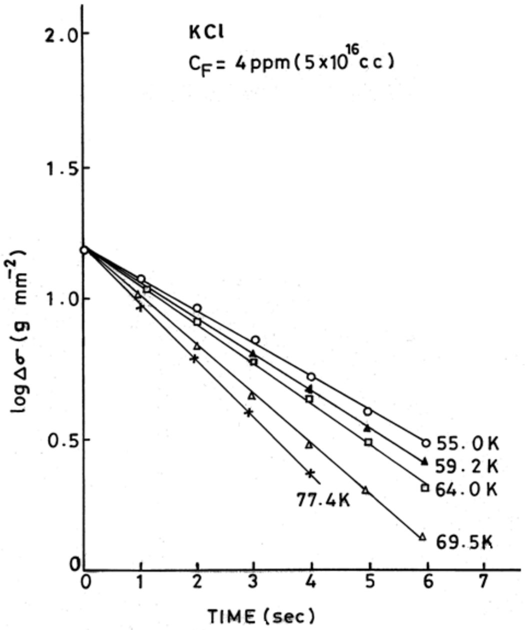

Fig. 11. Kinetics of the stress decay after light is turned off. Additively colored KCI[30].

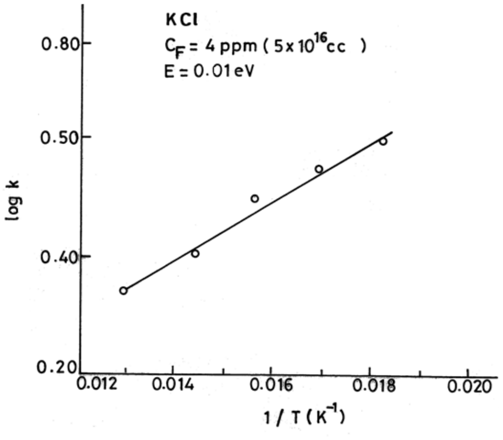

Fig. 12. Arrhenius plot of rate constant for stress decay in additively coloured KCI [30].

4. Conclusions

The important conclusions drawn from the study of photoplastic effect in coloured alkali halide crystals are as given below:

(i) Considering the basic process of PPE, an expression is derived for PPE, which is as given below

$$(\Delta\sigma)_s = \frac{D\delta_2 \dot{\varepsilon} r_i^0 \gamma_2 A n_{FO} I_L}{\delta_0 b\beta(\gamma_1+\gamma_2)(\delta-\alpha_e \dot{\varepsilon} t)} \exp(-\alpha_e \varepsilon) \left[\exp(-\alpha_e \varepsilon)-\exp\left(-\frac{\delta\varepsilon}{\dot{\varepsilon}}\right)\right]\left\{1-\exp[-\delta_0(t-t_c)]\right\}$$

(ii) The proposed theory is able to explain temporal, deformation, strain-rate, spectral, light intensity, coloration, dimension and thermal characteristic of PPE.

(iii) PPE can be used to determine the lifetime of the dislocation-captured F’ -cen tres in crystals and the pinning time of dislocations. It is shown that the lifetime of dislocation-captured F’-centres is longer as compared to the lifetime of normal F’-centres, i.e., the dislocation non-capture F’-centres present in the bulk.

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*Corresponding author: drsanjaytiwari@gmail.com