Theoretical approach to the Mechanoluminescence induced by elastic deformation of coloured alkali halide crystals using pressure steps

Raj kumar Goutam[1], Dr. G. Varma[1] and Ajay Kumar Baghel[2]

[1]M.H. college of Home Science and Science for women, Jabalpur, India

[2]Govt. P.G. College Lakhnadawa, Shivani, India

Abstract: The theoretical approach to the ML induced by the application of loads on coloured alkali halide crystals to make comparisons between the theoretical and experimental results. It is shown that the time constant for the rise of pressure and the pinning time of dislocations and the work hardening exponent can be determined from the measurement of the time dependence of ML and the pressure dependence of ML.

Keywords: Mechanoluminescence, Dislocation, coloured alkali halide crystals.

Introducuction

Most of the studies on the ML have been made either during slow deformation of the crystals or during fracture of the crystals. Only limited studies have been made on the ML induced during the application of loads on the crystals. Preliminary report of the ML emission during application of pressure on coloured alkali halide crystals were given by Urbach (1930) and Trink (1938). Kyuglov*et al.* (1966) have made a systematic study of the luminescence produced by plastic deformation of γ-irradiated KCl crystals from a stress lower than the elastic limit up to the breakdown stress of the crystals. They have reported that the luminescence of γ-irradiated KCl crystals induced by plastic deformation occurs during loading from a stress lower than the elastic limit to the breakdown stress of the crystal. A similarity between the stress-strain curve and the load luminescence curve is established. A luminescence yield stress of the irradiated crystals can be determined from the load-luminescence curves. Studies on slip band formation, pre-irradiation treatment of the crystals, and the effect of rate of load application have been presented. Qualitative information about the nature of the luminescence produced during the deformation is given. A tentative model of the luminescence phenomenon from the start of the elastic deformation upto the stress under which the crystals is heavily deformed has been reported. The model is based on the fact that the region around dislocations contains a higher concentration of colourcentres than the average concentration in the bulk. The instability introduced to the regions of high concentration of colourcentres produces a considerable amount of luminescence spikes in the luminescence produced at high stresses are due to the formation of slip bands.

Mechanism of ML Induced by Elastic Deformation of Coloured Alkali Halide Crystals Using Pressure-Steps

When a pressure is applied on to a crystal, initially the pressure increases with time and then it attains a final value P_o. If τ_r is the time–constant for rise of pressure P, then the increase of pressure with time may be given by the following equation

$$P = P_o\left[1-\exp(-t/\tau_r)\right] = P_o\left[1-\exp(-\xi t)\right] \quad \text{----} \quad (1)$$

where, $\xi = 1/\tau_r$, is the rate-constant for the rise of pressure with time.

If Y is the Young's modulus of elasticity of the crystal, then in the elastic region the strain ε produced in the crystal may be expressed as

$$\varepsilon = \frac{P_o}{Y}[1-\exp(-\xi t)] \quad \text{---} \quad (2)$$

where, P_o/Y is the maximum strain for a given pressure P_o.

From eq. (2), the strain rate $\dot{\varepsilon}$ may be expressed as

$$\dot{\varepsilon} = \frac{P_o}{Y}\xi \exp(-\xi t) \quad \text{---} \quad (3)$$

Considering that the rate of generation of bending segments of the dislocations is proportional to the strain rate, then we may write

$$\frac{dN_s}{dt} = G_s - \frac{N_s}{\tau_s} = \frac{BP_0}{Y}\xi \exp(-\xi t) - \delta N_s \quad \text{---} \quad (4)$$

where B is the proportionality constant, $\delta = 1/\tau_s$, and N_s is the number of bending dislocation segments at any time t.

Integrating eq. (4) and taking $N_s = 0$ at t = 0, we get

$$N_s = \frac{BP_0\xi}{Y(\xi-\delta)}\left[\exp(-\delta t) - \exp(-\xi t)\right] \quad \text{---} \quad (5)$$

If v_s is the average velocity of the bending of the dislocation segments, then the rate of area swept out by the dislocation segment may be expressed as

$$\frac{dS}{dt} = N_s v_s = \frac{BP_0\xi v_s}{Y(\xi-\delta)}\left[\exp(-\delta t) - \exp(-\xi t)\right] \quad \text{----} \quad (6)$$

As discussed previously the rate of generation G_i of the interacting F-centres and the generation of electrons in the dislocation band may be written as

$$G_i = \frac{n_F r_F BP_0\xi v_s}{Y(\xi-\delta)}\left[\exp(-\delta t) - \exp(-\xi t)\right] \quad \text{---} \quad (7)$$

If α_1 is the rate constant for jumping of the electrons from interacting centres to the dislocation band lying just above the F–centre level, and α_2 is the rate constant for the dropping back of the electrons from the interacting level (Chandra 1998), then we can write the following rate equation

$$\frac{dn_i}{dt} = G_i - \alpha_1 n_i - \alpha_2 n_i = \frac{n_F r_F BP_o \xi v_s}{Y(\xi - \delta)}\left[\exp(-\delta t) - \exp(-\xi t)\right] - \alpha n_i \quad \text{--- (8)}$$

where, $\alpha_1 = (\alpha_1 + \alpha_2)$, and $1/\alpha = \tau_i$, is the lifetime of interacting F–centres or the damping time of the segments of dislocations.

Integrating eq. (8) and taking $n_i = 0$, at $t = 0$, we get

$$n_i = \frac{BP_0 \xi n_F r_F v_s}{Y(\xi - \delta)\alpha}\left[\frac{\exp(-\delta t)}{(\alpha - \xi)} - \frac{\exp(-\xi t)}{(\alpha - \xi)} + \frac{(\xi - \delta)}{(\alpha - \xi)(\alpha - \delta)}\exp(-\alpha t)\right] \quad \text{--- (9)}$$

For $\alpha \gg \xi$, and $\alpha \gg \delta$, we get

$$n_i = \frac{BP_0 \xi n_F r_F v_s}{Y(\xi - \delta)\alpha}\left[\exp(-\delta t) - \exp(-\xi t)\right] \quad \text{--- (10)}$$

Thus, the rate of generation of G_d of electron in the dislocation band is given by

$$G_d = \alpha_1 n_i = \frac{\alpha_1 BP_0 \xi n_F r_F v_s}{Y(\xi - \delta)\alpha}\left[\exp(-\delta t) - \exp(-\xi t)\right]$$

or, $G_d = G_0\left[\exp(-\delta t) - \exp(-\xi t)\right]$ --- (11)

where, $G_0 = \dfrac{BP_0 \xi\, p_F n_F r_F v_s}{Y(\xi - \delta)}$ and $p_F = \alpha_1/\alpha$, is the efficiency of the capture of interacting F- centre electron by the dislocation segments.

In X or γ–irradiated alkali halide crystals, the ML emission takes place due to the movement of electrons with dislocation segments as well as due to the movement of electrons along the dislocation axis. Suppose σ_1, σ_2 and σ_3 and $\sigma`_1$, $\sigma`_2$ and $\sigma`_3$ are the capture cross–sections of the hole centres, negative ion vacancies and other traps for the electrons moving with velocity v_s together with the dislocation segments and electrons moving with velocity v_e along the dislocation axis, respectively, and if N_1, N_2 and N_3 are the densities of hole centres, negative ion vacancies and other centres, respectively, then we can write the following rate equation

$$\frac{dn_d}{dt} = G_d - (\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3)v_s n_d - (\sigma_1' N_1 + \sigma_2' N_2 + \sigma_3' N_3)v_e n_d$$

or,

$$\frac{dn_d}{dt} = G_o\left[\exp(-\delta t) - \exp(-\beta t)\right] - (\beta_1 + \beta_2 + \beta_3)n_d - (\beta_1' + \beta_2' + \beta_3')n_d$$

$$\frac{dn_d}{dt} = G_o\left[\exp(-\delta t) - \exp(-\beta t)\right] - (\beta + \beta')n_d = g_o\left[\exp(-\delta t) - \exp(-\beta t)\right] - \beta_o n_d \quad \text{----(12)}$$

Here, $\beta_1 = \sigma_1 N_1 v_s$, $\beta_2 = \sigma_2 N_2 v_s$, $\beta_3 = \sigma_3 N_3 v_s$, $\beta`_1 = \sigma`_1 N_1 v_e$, $\beta`_2 = \sigma`_2 N_2 v_e$, $\beta`_3 = \sigma`_3 N_3 v_e$, $\beta = (\beta_1 + \beta_2 + \beta_3)$, $\beta` = (\beta`_1 + \beta`_2 + \beta`_3)$ and $\beta_o = \beta + \beta`$

Integrating eq. (12) and taking $n_d = 0$, at $t = 0$, we get

$$n_d = G_o\left[\frac{\exp(-\delta t)}{(\beta_o-\delta)} - \frac{\exp(-\xi t)}{(\beta_o-\xi)} - \frac{(\xi-\delta)\exp(-\beta_o t)}{(\beta_o-\delta)(\beta_o-\xi)}\right] \quad --- \quad (13)$$

When the pressure applied on to the crystals increases slowly, then dP/dt or dε/dt is less. Therefore, in this case, $\beta_o >> \xi$, and $\beta_o >> \delta$, and eq.(13) may be expressed as

$$n_d = \frac{G_o}{\beta_o}\left[\exp(-\delta t) - \exp(-\xi t)\right] \quad --- \quad (14)$$

If η is the efficiency of radiative electron–hole recombination, then the ML intensity may be expressed as

$$I = \eta(\beta_1+\beta_1')n_d = \frac{\eta(\beta_1+\beta_1')G_o}{(\beta+\beta')}\left[\exp(-\delta t) - \exp(-\xi t)\right] \quad ---- \quad (15)$$

For $v_s << v_e$, $\beta << \beta'$, and $\beta_1 << \beta'_1$, and thus eq. (15) may be written as

$$I = \frac{\eta\beta_1' G_o}{\beta'}\left[\exp(-\delta t) - \exp(-\xi t)\right] \quad --- \quad (16)$$

(i) **Rise of ML intensity**

From eqs. (16) rise of ML intensity may be expressed as

$$I_r = \frac{\eta\beta_1' G_0}{\beta_1}(\xi-\delta)t$$

$$I_r = \frac{\eta\beta_1' BP_0 \xi p_F r_F n_F v_s t}{\beta_1 Y} \quad --- \quad (17)$$

Equation (17) indicates that initially the ML intensity should increase linearly with time t.

(ii) **Estimation of t_m, I_m, and I_T**

Equation (16) indicates that the ML intensity will be maximum at a time t_m given by

$$t_m = \frac{1}{(\xi-\delta)}\ln\left[\frac{\xi}{\delta}\right] \quad --- \quad (18)$$

From eqs. (16) and (18), the maximum ML intensity I_m may be expressed as

or, $$I_m = \frac{\eta\beta_1' p_F n_F r_F BP_0 \xi v_s}{\beta' Y \delta}\left(\frac{\delta}{\xi}\right)^{\frac{1}{(1-\delta/\xi)}} \quad --- \quad (19)$$

For $\xi >> \delta$, eq. (19) may be expressed as

$$I_m = \frac{\eta\beta_1' p_F n_F r_F BP_0 v_s}{\beta' Y} \quad --- \quad (20)$$

Using eq. (16), the total integrated ML intensity I_T, may be given by

$$I_T = \int_o^\infty I dt = \eta \frac{\beta_1'}{\beta} \frac{p_F n_F r_F BP_o v_s}{Y\delta} \quad --- \quad (21)$$

(iii) **Decay of ML Intensity**

Equation (16) may be written as

$$I = \frac{\eta \beta_1' G_o}{\beta'} \exp(-\delta t)[1 - \exp\{-(\xi - \delta)t\}]$$

or,
$$I = \frac{\eta \beta_1' G_o}{\beta'} \exp(-\delta t_m)\exp[-\delta(t - t_m)][1 - \exp\{-(\xi - \delta)t\}]$$

or,
$$\frac{I}{[1 - \exp\{-(\xi - \delta)t\}]} = \frac{\eta \beta_1' G_o}{\beta'} \exp(-\delta t_m)\exp[-\delta(t - t_m)] \quad ---- \quad (22)$$

for $(\xi - \delta)t >> 1$,eq.(22) may be expressed as

$$I = \frac{\eta \beta_1' G_o}{\beta'} \exp(-\delta t_m)\exp[-\delta(t - t_m)]$$

or,
$$I_d = I_m' \exp[-\delta(t - t_m)] \quad --- \quad (23)$$

where $I_m' = \frac{\eta \beta_1' G_o}{\beta'} \exp(-\delta t_m)$, is the interpolated value of I, at $t = t_m$.

The above equation indicates that the exponential decay of ML intensity with time where the decay time will be equal to the damping time of the dislocation segments. Thus, $\delta = 1/\tau_r$, can be determined from the slope of ln I versus $(t - t_m)$ plot, and if δ and t_m are known, ξ can be evaluated using equation (18).

(iv) **Pressure dependence of t_m,I_m and I_T**

As $\xi >> \delta$, and δ increase with pressure, eq. (18) indicates that t_m should decrease slightly with increasing pressure. As the both δ and v_s increases with the strain rate $\dot{\varepsilon}$, we may take $v_s = D\delta$, where D is a constant. Thus, eqs.(20) and (21) may be expressed as

$$I_m = \frac{\eta \beta_1' p_F n_F r_F BP_0 D\delta}{\beta' Y} \quad --- \quad (24)$$

and,
$$I_T = \frac{\eta \beta_1' p_F n_F r_F BDP_0}{\beta' Y} \quad --- \quad (25)$$

Equation (24) indicates that I_m/δ should increases linearly with P_0, and eq. (25) indicates that I_T should increases linearly with P_0.

(v) **Coloration dependence of I_m and I_T**

It is evident from eqs. (24) and (25) that both I_m and I_T should increases linearly with the density of F-centres.

(vi) **Temperature Dependence of t_m, I_m and I_T**

The efficiency p_F increases with temperature and it may be expressed as (Chandra 1998)

$$p_F = p_F^o \exp\left(-\frac{E_a}{kT}\right) \quad --- \quad (26)$$

where, E_a is the activation energy and p_F^o is a constant.

From eqs. (24), (25) and (26), we get

$$I_m = \frac{\eta \beta_1' p_F^o n_F r_F BD\delta P_o \exp(-E_a/kT)}{\beta' Y} \quad --- \quad (27)$$

and,

$$I_T = \frac{\eta \beta_1' p_F^o n_F r_F BDP_o \exp(-E_a/kT)}{\beta' Y} \quad --- \quad (28)$$

At low temperature there will be no thermal bleaching, and both I_m and I_T at a given pressure should increase with increasing of the crystals temperature and the dependence of I_m/δ on T and I_T on T should follow the Arrhenius plot. At higher temperature, n_F will decrease with increasing temperature because of thermal bleaching and consequently both I_m and I_T should decrease with increasing temperature. Thus, both I_m and I_T should be optimum for a particular temperature of the coloured alkali halide crystals.]

Experimental Support on the Proposed Theory

Fig **1** shows that when a pressure step (load) of small amplitude below the limit of elasticity is applied on to γ-irradiate KBr crystal, then initially the ML intensity increases with time attain of peak value and later on it decreases with time. Such results are expected from eq.(16). Fig.**2** the plots of log I_D versus (t-t_m) are straight lines and there is not-linearity for smaller value of ($t - t_m$). This is in accordance with eq. (22). Fig **3** shows that the plot of log I/ [1-exp {-(ξ-δ)}] versus ($t - t_m$) are straight lines with negative slope. This is in accordance with eq.(22). Fig. **4** illustrates the value of t_m, τ_r and τ_s for different pressures. It seems that where asτ_s and t_m decreases with increasing pressure P_0, $\tau_r$does not change significantly with increasing P_0. Fig. **5** shows that $I_m/\xi(\delta/\xi)^{1-\delta/\xi}$ and I_T increases linearly with the applied pressure P_o. These results follows eq.(19) and (21). **Table 1** shows the value of t_m, τ_r, τ_s and ξ for different pressure.

Table 1: Values of t_m, τ_r, δ, ξ, τ_s for the ML of γ-irradiated KBr crystal

Pressure g/mm^2	**t_m(s)**	**δ (s^{-1})**	**ξ (s^{-1})**	**τ_s (s)**	**τ_r (s)**
10	**0.14**	**4.887**	**10.101**	**0.205**	**0.099**
20	**0.13**	**5.370**	**10.500**	**0.186**	**0.095**
30	**0.12**	**6.152**	**10.801**	**0.163**	**0.093**

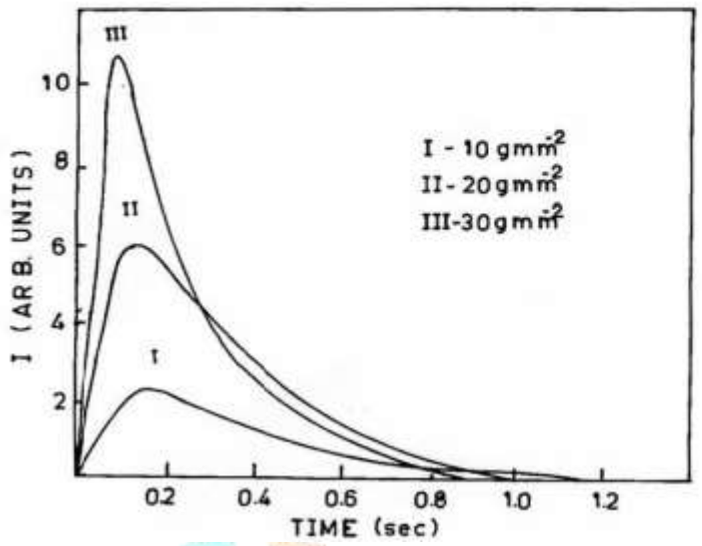

Figure 1 Time dependant of the ML intensity of γ-irradiated KBr crystals induced by elastic deformation produced during the application of pressure steps.

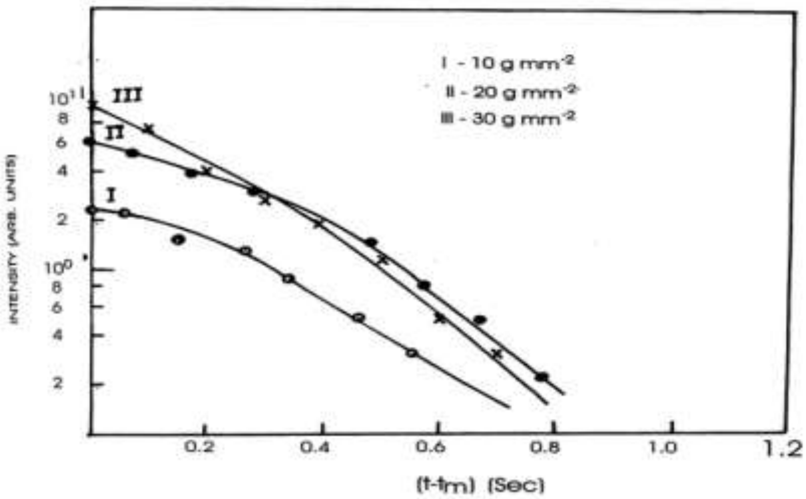

Figure 2 Plot of log I_d versus $(t-t_m)$ for γ-irradiated KBr crystals.

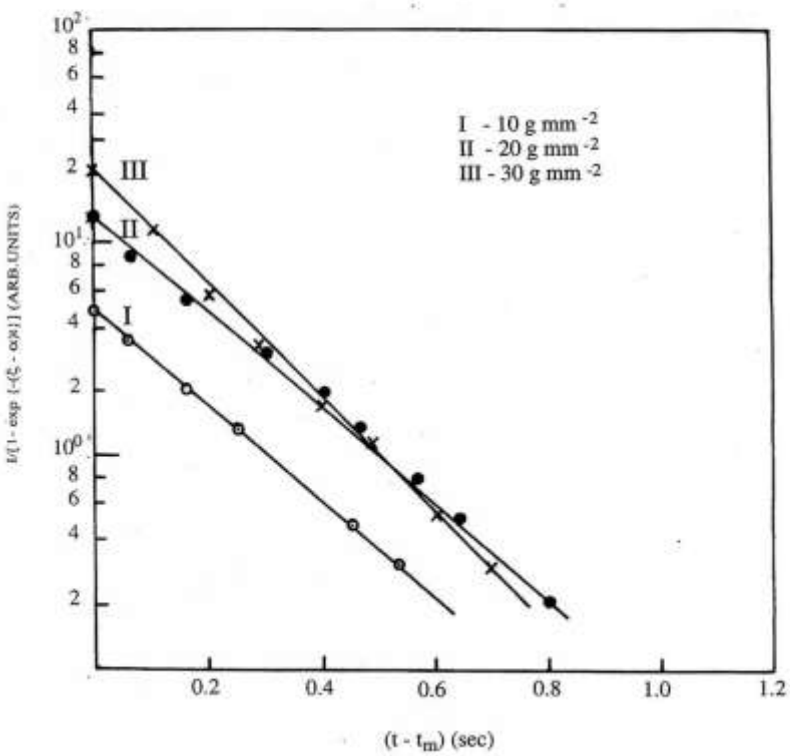

Figure 3 Plot of $[I_m/\{\xi(\delta/\xi)^{1/(1-\alpha/\xi)}\}]$ versus $(t-t_m)$.

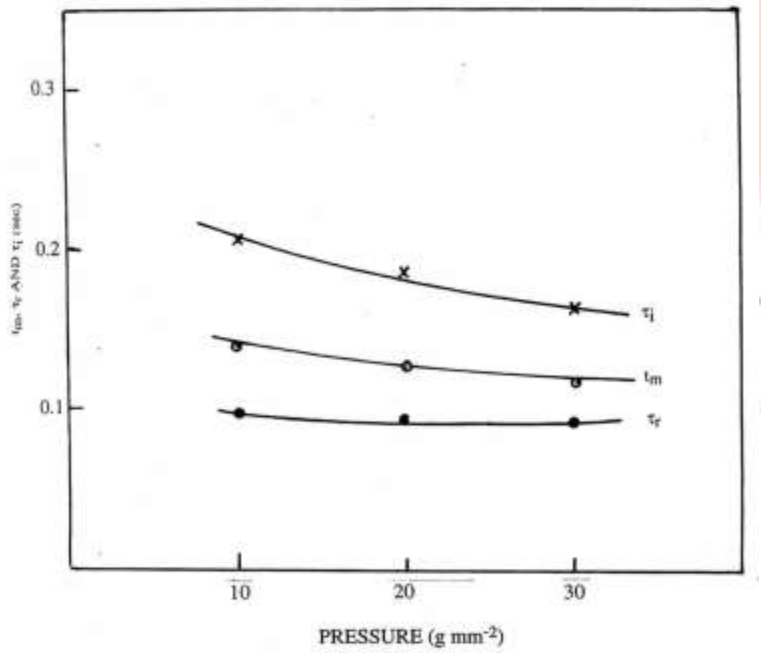

Figure 4 Dependence of t_m, τ_r, and τ_i of γ-irradiated KBr crystals on the applied pressure P_0.

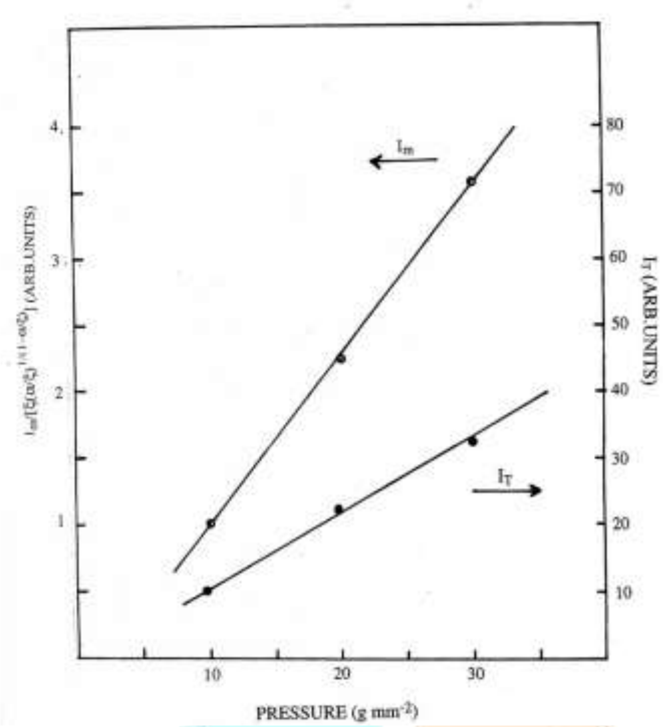

Figure 5 Dependence of [$I_m/\{\xi(\delta/\xi)^{1/(1-\alpha/\xi)}\}$] and I_T of γ-irradiated KBr crystals on the applied pressure P_0.

Conclusions

The important conclusions drawn from the studies of the ML induced by application of pressure steps oncoloured alkali halide crystals are the ML in X or γ-irradiated alkali halide crystals can be induced by applying uniaxial pressure much below the critical stress for the limit of elasticity. When a uniaxial pressure much below the elasticity limit is applied on to γ-irradiated alkali halide crystals, then initially the ML intensity increases with time, attains a peak value and later on it decreases exponentially with time.In coloured alkali halide crystals the values of both I_m and I_T increase with pressure and also with the density of F-centres in the crystals. Both I_m and I_T are optimum for a particular temperature of the crystals.From the measurement of the time dependence of ML intensity in coloured alkali halide crystals, the value of rise time of τ_r of pressure, damping time of dislocation segments, pinning time of dislocations, and the work hardening exponent can be determined.

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