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Mechanoluminescence and Deformation Bleaching in Coloured Alkali Halide Crystals

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The present paper reports the mechanoluminescence (ML) and deformation bleaching of a colouration in coloured alkali halide crystals. As the F-centre electrons captured by moving dislocations are picked up by holes, deep traps and other compatible traps, so deformation bleaching occurs. Simultaneously radiative recombination of dislocation captured electrons with the holes gives rise to the mechanoluminescence. Expressions have been derived for the strain dependence of the density of colour centres is deformed crystals and for the number of colour centres bleached. So far as strain, temperature, density of colour centres, E_a and volume dependence are concerned, there exists a correlation between the deformation bleaching and mL in coloured alkali halide crystals. From the strain dependence of the density of colour centres in deformed crystals, the value of coefficient of deformation bleaching D is found to be 1.929 and 1.990 for KCl and KBr crystals respectively. The value of $(D + \chi)$ as determined from the strain dependence of the ML intensity is found to be 2.59 and 3.69 for KCl and KBr crystals respectively. This gives the value of coefficient of deformation generated compatible traps χ to be 0.68 and 1.69 for KCl and KBr crystals respectively.

INTRODUCTION

The light emission produced during deformation (the phenomenon of luminescence) has been reported in alkali halide crystals by a number of authors summarized in tabular form as follows:

	Authors	Contributions
1.	Urbach[1]	The phenomenon of luminescence in alkali halide crystals.
2.	Wick[2]	ML emission during deformation of coloured alkali halide crystals.
3.	Trinks[3]	Increase in ML intensity of NaCl and KCl crystals with irradiation doses, thickness of the crystals and with pressure.
4.	Metz *et al.*[4]	The linear dependence of light emission on the strain rate in X-ray irradiatd KBr, NaCl and LiF crystals.

5.	Pirog *et al.*[5]	The decrease in ML intensity with increasing rate of compression.
6.	Senchukov *et al.*[6]	ML occurs in most of the cases due to the recombination of free electrons with luminescence centres.
7.	Chandra *et al.*[7]	The dislocation movement to be responsible for the ML excitation in coloured alkali halide crystals.
8.	Butler[8]	The ML spectra of γ-irradiated alkali halide crystals are similar to luminescence excited by high energy radiation.
9.	Ueta *et al.*[9]	The decay curve of the electric current produced during plastic deformations of nonirradiated KCl crystals and the decay curve of the ML produced during deformation of the irradiated crystals are of the same form.
10.	Chandra *et al.*[10]	The ML is produced in coloured alkali halide crystals during the application of pressure as well as during the release of applied pressure.
11.	Guerrero *et al.*[11]	The dependence of ML and thermoluminescence on the strain of irradiated KCl crystals.
12.	Hordy *et al.*[12]	The ML excited by 1060 nm Na glass laser beam is similar in spectra to the ML excited by plastic deformations of χ- or γ-irradiated alkali halide crystals.
13.	Mayer *et al.*[13]	The ML and thermoluminescence in γ- irradiated KCl crystals.
14.	Miyake *et al.*[14]	The effect of annealing in chlorine gas on the ML of X-rays irradiated KCl crystals.
15.	Ossipyan *et al.*[15] and Molotski *et al.*[16, 17]	The mechanism of ML excitation in coloured alkali halide crystals.
16.	Hagihara *et al.*[18]	Process of ML excitation in γ-irradiated KCl crystals.
17.	Atari,[19]	Dependence of the mL of coloured alkali halide crystals on different parameters.
18.	Zakrevski *et al.*[20]	The electron emission and luminescence associated with the plastic deformation of ionic crystals.
19.	Chandra[21]	The dependence of ML of coloured alkali halide crystals on different parameters.
20.	Chandra *et al.*[22]	Post-irradiation deformation causes deformation bleaching in coloured alkali halide crystals.

As bleaching of colouration and the mL emission is caused by the post-irradiation deformation, a correlation between the deformation bleaching and ML of coloured alkali halide crystals is expected. In this article the author has attempted to correlate the deformation bleaching and mL of coloured alkali halide crystals.

Dependence of Deformation Bleaching on Parameters

If a coloured alkali halide crystal is plastically deformed, the movement of dislocation takes place. These moving dislocations may capture electrons from the colour centres and may subsequently transport the captured electrons to hole centres, deep traps and other compatible traps in the crystal and thereby deformation bleaching of the colouration in alkali halide crystals may take place. Let a crystal contain Na dislocations of unit length/unit volume. As Na dislocations move through a distance dx, the area swept out by the dislocations will be Na dx. The deformation bleaching in coloured alkali halide crystals may take place due to the transfer of electrons from F-centres to the dislocation band and their subsequent recombination with other centres.

The density η_F of the F-centres in coloured alkali halide crystals decreases with post-irradiation deformation of the crystals. The dependence of η_F on the strain ε may be expressed as

$$\eta_F = \eta_{FO} \exp(-D\varepsilon) \qquad (1)$$

where D is the coefficient of deformation bleaching given by

$$D = p_F r_F / b$$

where r_F is the distance up to which a dislocation can interact with colour centre, p_F is the dislocation capture-probability of F-centre electrons and b is Burger's vector. η_{FO} is the density of F-centres in the undeformed crystals.

For a crystal of volume V

$$\eta'_F = \eta_F V = \eta_{FO} V \exp(-D\varepsilon) \qquad (2)$$

Hence the number of colour centres bleached in a crystal of volume V at deformation ε may be expressed as

$$\Delta\eta_F = \eta_{FO} V[1 - \exp(-D\varepsilon)] \qquad (3)$$

Dependence of mL on Parameters

In case when alkali halide crystal is deformed, the moving dislocations capture electrons from the nearby F-centres as the dislocation band lies just above the ground state of F-centre level[16, 17]. The rate of generation of electrons in the dislocation band may be expressed by

$$g = \dot{\varepsilon}/b p_F r_F \eta_F \qquad (4)$$

where $\dot{\varepsilon}$ is the strain rate of the crystal.
From equations (1) and (2) we have

$$g = (p_F \eta_{FO} r_F \dot{\varepsilon}/b) \exp(-D\varepsilon)$$

or

$$g = g_0 \exp(-D\varepsilon) \qquad (5)$$

where $g_0 = p_F \eta_{FO} r_F \dot{\varepsilon}/b$

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

$$d\eta_d/dt = g - \sigma_1 N_1 v_d n_d - \sigma_2 N_2 v_d n_d - \sigma_3 N_3 v_d n_d \tag{6}$$

where n_d = number of electrons in the dislocation band at time t.

σ_1, σ_2 and σ_3 = the cross-sections and N_1, N_2 and N_3 = densities of hole centres, deep traps and other compatible traps respectively.

v_d = velocity of dislocations.

The velocity of electrons has been taken as the velocity of dislocations because the dislocation captured electrons move with the dislocations. Here, the compatible traps means the traps whose electron capture-probability is much greater than that of 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 process.

From equations (5) and (6) we find

$$dn_d/dt = g_0 \exp(-D\varepsilon) - \gamma n_d \tag{7}$$

or

$$dn_d/dt = g_0 \exp(-D\dot{\varepsilon}t) - \gamma n_d$$

where $\gamma = 1/T_d = (\sigma_1 N_1 - \sigma_2 N_2 + \sigma_3 N_3) v_d$ (8)

and T_d is the life time of electrons in the dislocation band.

Integrating equation (7) and taking $n_d = 0$ at $t = 0$, we have

$$n_d = g_0/\gamma - D\dot{\varepsilon}\,[\exp(-D\dot{\varepsilon}) - \exp(-\gamma t)] \tag{9}$$

From equation (6), the rate of recombination of dislocation electrons with holes may be given by

$$R_h = \sigma_1 N_1 v_d n_d$$

or

$$R_h = \sigma_1 N_1 v_d g_0/(\gamma - D\dot{\varepsilon})[\exp(-D\dot{\varepsilon}t) - \exp(-\gamma t)] \tag{10}$$

Substituting v_d from equation (8), we get

$$R_h = \frac{\sigma_1 N_1 g_0 \gamma}{(\sigma_1 N_1 + \sigma_2 N_2 + \sigma_3 N_3)(\gamma - D\dot{\varepsilon})}[\exp(-D\dot{\varepsilon}t) - \exp(-\gamma t)] \tag{11}$$

As the number of deep traps is less as compared to other centres, we have

$$R_h = \frac{\sigma_1 N_1 g_0 \gamma}{(\sigma_1 N_1 + \sigma_3)(\gamma - D\dot{\varepsilon})}[\exp(-D\dot{\varepsilon}t) - \exp(-\gamma t)]$$

or

$$R_h = \frac{g_0 \gamma}{(1 + (\sigma_3 N_3/\sigma_1 N_1))(\gamma - D\dot{\varepsilon})}[\exp(-D\dot{\varepsilon}t) - \exp(-\gamma t)] \tag{12}$$

The electron-traps are created by the plastic deformation of alkali halide crystals whose density increases more or less linearly with the deformation of crystals[23]. Thus the dependence of the number N_ε of newly created electron traps due to the strain may be expressed as

$$N_{\dot{\varepsilon}} = M\dot{\varepsilon}$$

where M is the multiplication factor.

Out of the deformation generated N_ε electron traps, a fraction A of them may have electron capture-probability greater than that of dislocations and thus the number of deformation generated compatible electron traps N_3 may be given by

$$N_3 = AM\dot{\varepsilon} \tag{13}$$

Thus equation (12) may be written as

$$R_h = g_0\gamma/(1 + \chi\dot{\varepsilon})(\chi - D\dot{\varepsilon})\ [\exp(-D\dot{\varepsilon}t) - \exp(-\gamma t)] \tag{14}$$

where $\chi = (\sigma_3 AM/\sigma_1 N_1)$ is the coefficient of deformation generated compatible traps.

When $\chi\dot{\varepsilon}$ is less than 1, equation (14) may be written as

$$R_h = [g_0\gamma/(\gamma - D\dot{\varepsilon})]\exp(-\chi\dot{\varepsilon})[\exp(-D\dot{\varepsilon}t) - \exp(-\gamma t)] \tag{15}$$

The probability η of the radiative recombination of electrons with hole centres may be assumed to be constant for low deformation. Thus, the ML intensity may be expressed as

$$I = \eta R_h$$

or

$$I = [\eta g_0\gamma/(\gamma - D\dot{\varepsilon})]\exp(-\chi\dot{\varepsilon})\ [\exp(-D\dot{\varepsilon}t) - \exp(-\gamma t)] \tag{16}$$

Putting the value of g_0, we get

$$I = [\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}/(\gamma - D\dot{\varepsilon})b]\exp(-\chi\dot{\varepsilon})[\exp(-D\dot{\varepsilon}t) - \exp(-\gamma t)]$$

or

$$I = [\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}/(\gamma - D\dot{\varepsilon})b]\{\exp[-(D + \chi)\dot{\varepsilon}] - \exp[-\gamma/\dot{\varepsilon} + \chi]\dot{\varepsilon}\} \tag{17}$$

For low value of $\dot{\varepsilon}$,

$$I = [\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}/(\gamma - D\dot{\varepsilon})b][\gamma/\dot{\varepsilon} + \chi - D - \chi]\dot{\varepsilon}$$

or

$$I = [\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}/b] \tag{18}$$

Equation (18) indicates that for low value of ε, the ML intensity should increase linearly with the strain $\dot{\varepsilon}$.

Equation (17) shows that $I = 0$ at $\dot{\varepsilon} = 0$ and $I = 0$ at $\dot{\varepsilon} = \infty$. Thus, the ML intensity should be maximum for a particular value of the strain. By equating $dI/d\dot{\varepsilon} = 0$, we get the value of strain. $\dot{\varepsilon}_m$ at which ML intensity will be maximum. From equation (17), we get

$$(D + \chi)\exp[-(D + \chi)\dot{\varepsilon}] = (\chi + (\gamma/\dot{\varepsilon}))\exp[-(\chi + (\gamma/\dot{\varepsilon})\dot{\varepsilon}]$$

or

$$\exp[-(D + \chi)\dot{\varepsilon}] = (\chi + (\gamma/\dot{\varepsilon})/(D + \chi)\exp[-(\chi + (\gamma/\dot{\varepsilon})\dot{\varepsilon}]$$

Put $\dot{\varepsilon} = \dot{\varepsilon}_m$,

$$\exp[\chi + (\gamma/\dot{\varepsilon}) - D - \chi]\dot{\varepsilon}_m = (\chi + (\gamma/\dot{\varepsilon}))/(D + \chi)$$

or

$$\dot{\varepsilon}_m = 1/[(\gamma/\dot{\varepsilon}) - D]\ \mathrm{In}\,(\chi + (\gamma/\dot{\varepsilon}))/(D + \chi) \tag{20}$$

From equations (17) and (19) we get the maximum value of $I = I_m$ as

$$I_m = \frac{\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}}{(\gamma - D\dot{\varepsilon})b}\{\exp[-(\chi + (\gamma/\dot{\varepsilon}))\dot{\varepsilon}_m][\chi + (\gamma/\dot{\varepsilon})/(D + \chi) - 1]\} \tag{21}$$

Substituting the value of $\dot{\varepsilon}_m$ from equation (20), we get

$$\mathrm{Im} = \frac{\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}}{(\gamma - D\dot{\varepsilon})b}\left\{\exp\left[-\frac{(\chi + (\gamma/\dot{\varepsilon}))}{(\gamma/\dot{\varepsilon} - D)}\,\mathrm{In}\,\frac{(\chi + (\gamma/\dot{\varepsilon}))}{(D+\chi)}\,\frac{(\chi + (\gamma/\dot{\varepsilon}))}{(D+\chi)} - 1\right]\right\} \tag{22}$$

For $\gamma \gg D\dot{\varepsilon}$ and $\gamma \gg \chi\dot{\varepsilon}$,

$$\mathrm{Im} = \frac{\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}}{b}\exp\left[\left\{\frac{(D+\chi)}{(\chi + (\gamma/\dot{\varepsilon}))}\,\frac{(\chi + (\gamma/\dot{\varepsilon}) - D - \chi)}{(D+\chi)}\right\}\right]$$

or

$$\mathrm{Im} = \frac{\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}}{b} \tag{23}$$

As $\gamma \gg \chi\dot{\varepsilon}$ for large value of $\dot{\varepsilon}$ equation (17) may be expressed as

$$I = \frac{\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}}{b}\exp\left[-(D+\chi)\dot{\varepsilon}\right] \tag{24}$$

Equation (24) indicates that for large deformation, I should decrease exponentially with $\dot{\varepsilon}$.

From equation (17) the total ML intensity I_T, *i.e.*, the total number of photons emitted up to the strain $\dot{\varepsilon}$ of the crystal may be expressed as

$$I_T = \int_0^{\dot{\varepsilon}} I\,d\dot{\varepsilon}$$

or

$$I_T = \frac{\eta\gamma p_F\eta_{FO}r_F\dot{\varepsilon}}{(\gamma - D\dot{\varepsilon})b}\int_0^{\dot{\varepsilon}}\{\exp\left[-(D+\chi)\dot{\varepsilon}\right] - \exp\left[-((\gamma/\dot{\varepsilon}) + \chi)\dot{\varepsilon}\right]\}d\dot{\varepsilon}$$

As $\gamma \gg D\dot{\varepsilon}$, we get

$$I_T = \frac{\eta p_F\eta_{FO}r_F\dot{\varepsilon}}{b}\int_0^{\dot{\varepsilon}}\{\exp\left[-(D+\chi)\dot{\varepsilon}\right] - \exp\left[-((\gamma/\dot{\varepsilon}) + \chi)\dot{\varepsilon}\right]\}d\dot{\varepsilon}$$

or

$$I_T = \frac{\eta p_F\eta_{FO}r_F\dot{\varepsilon}}{b}\left\{\frac{\exp\left[-(D+\chi)\dot{\varepsilon}\right]}{(D+\chi)}\right\}_0^{\dot{\varepsilon}} - \left\{\frac{\exp\left[-(\chi + (\gamma/\dot{\varepsilon}))\dot{\varepsilon}\right]}{-\chi - \gamma/\dot{\varepsilon}}\right\}_0^{\dot{\varepsilon}} \tag{25}$$

As $(\chi + (\gamma/\dot{\varepsilon})) \gg (D+\chi)$, the second term on R.H.S. of the equation may be neglected.

$$I_T = \frac{\eta p_F\eta_{FO}r_F\dot{\varepsilon}}{b(D+\chi)}\{1 - \exp\left[-(D+\chi)\dot{\varepsilon}\right\}$$

or

$$I_T = I_T^0\{1 - \exp\left[-(D+\chi)\dot{\varepsilon}\right]\} \tag{26}$$

where $I_T^0 = \eta p_F\eta_{FO}r_F\dot{\varepsilon}/b(D+\chi)$

Equation (26) indicates that the total number of photons emitted should initially increase linearly with the deformation of crystals and then it should attain a saturation value for large deformation.

The probability p_F of the transfer of electrons from an F-centre to the interacting dislocation is related to the transfer of electrons from the interacting F-centres to the dislocation band and its temperature dependence may be given as

$$p_F = p_F^0 \exp[-E_a/kT] \tag{27}$$

where E_a is the energy gap between the bottom of dislocation band and average ground state energy of the interacting F-centres and p_F^0 is a constant.

From equations (20), (26) and (27), we get

$$I_m = \eta \eta_{FO} r_F \dot{\varepsilon} p_F^0 / b \exp[-E_a/kT] \tag{28}$$

and

$$I_T = \eta \eta_{FO} r_F \dot{\varepsilon} p_F^0 / b(D + \chi) \exp[-E_a/kT]\{1 - \exp[-(D + \chi)\dot{\varepsilon}] \tag{29}$$

Equations (28) and (29) show that for given values of η_{FO} and $\dot{\varepsilon}$, both I_m and I_T should increase with temperature of the crystal. However, at higher temperature η_{FO} will decrease due to the thermal blcaching and I_m and I_T should be optimum for a particular temperature of the crystals.

Deformation bleaching and ML correlation in coloured alkali halide crystals

When the F-centre electrons captured by moving dislocations are picked up by holes, deep traps and other compatible traps, then deformation bleaching occurs. At the same time radiative recombination of dislocation captured electrons with the holes gives rise to the mechanoluminescence. Thus, there should be a correlation betweeen deformation bleaching and ML. In this context the following aspects are of particular interest:

1. From equation (3) deformation bleaching may be given by

   $$\Delta\eta_F = \eta_{FO} V[1 - \exp(-D\dot{\varepsilon})].$$

   The total number of photons emitted may by given by equation (26).

   $$I_T = I_T^0 [1 - \exp[-(D + \chi)\dot{\varepsilon}]]$$

   Thus, the strain dependence of deformation bleaching is slower as compared to the strain dependence of total ML intensity. From the strain dependence of deformation bleaching, the coefficient of deformation bleaching D may be calculated and using this value of D, the coefficient of deformation generated compatible traps χ may be determined from the strain dependence of I_T.
2. Whereas the ML intensity depends linearly on the strain rate $\dot{\varepsilon}$ (equation 18) the deformation bleaching weakly depends on the $\dot{\varepsilon}$ depending on the strain rate dependence of D (equation 1).
3. Both the deformation bleaching and ML should initially increase with the increasing temperature of the crystal because of the increase in the dislocation capture probability of F-centre electrons and both of them should decrease at high temperature because of the thermal bleaching of the coloration in alkali halide crystals. Hence, both the deformation bleaching and mechanoluminesence intensity should be optimum for a particular temperature of the crystals.
4. Both the deformation bleaching and ML should depend linearly on the density of colour centres in the undeformed crystals.
5. Because both the deformation bleaching and ML depend on the dislocation

capture probability of F-centre electrons, which decreases with the increasing value of E_a, *i.e.*, the energy gap between the dislocation band and ground state of F-centre electrons, they should consequently decrease with increasing value of E_a.

6. Both the deformation bleaching and ML intensity should increase with the volume V of the crystals.

Conclusion

When the F-centre electrons captured by moving dislocations are picked up by holes, deep traps and other compatible traps, thin deformation bleaching occurs. At the same time, radiative recombination of dislocation captured electrons with the holes gives rise to the mechanoluminescence. Thus, there should be correlation between deformation bleaching and ML.

Expressions are derived for the strain dependence of the density of colour centres in deformed crystals and also for the number of colour centres bleached given respectively below:

$$\eta_F = \eta_{FO} V \exp(-D\dot{\varepsilon})$$

and

$$\Delta\eta_F = \eta_{FO} V [1 - \exp(-D\dot{\varepsilon})]$$

The $\dot{\varepsilon}_m$, I_m, I_T and I for the effect of post-irradiation deformation on the ML intensity are derived as

$$\dot{\varepsilon}_m = 1/[(\gamma/\dot{\varepsilon}) - D] \mathrm{In} (\chi + (\gamma/\dot{\varepsilon}))/(D + \chi)$$

$$I_m = \{\eta\eta_{FO} r_F \dot{\varepsilon} p_F^0/b\} \exp[-E_a/kT]$$

$$I_T = [\eta\eta_{FO} r_F \dot{\varepsilon} p_F^0/b(D + \chi)] \exp[-E_a/kT]\{1 - \exp[-(D + \chi)\dot{\varepsilon}]$$

$$I = \frac{\eta p_F \eta_{FO} r_F \dot{\varepsilon}}{b} \exp[-(D + \chi)\dot{\varepsilon}]$$

From the strain dependence of the density of colour centres in reformed crystals, the value of coefficient of deformation bleaching D is determined and it is found to be 1.929 and 1.990 for KCl and KBr crystals respectively. The value of $(D + \chi)$ is determined from the strain dependence of the ML intensity and it is found to be 2.59 and 3.69 for KCl and KBr crystals respectively. This gives the value of coefficient of deformation generated compatible traps χ to be 0.68 and 1.69 for KCl and KBr crystals respectively.

So far as the strain, temperature, density of colour centres, E_a and volume dependences are concerned, there exists a correlation between the deformation bleaching and ML in coloured alkali halide crystals.

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