

Theory of Laser ML in Coloured Alkali Halide Crystals and II-VI Semiconductors

Manas Kumar Sahu

Dept. of Physics of Sound, Indira Kala Sangit Vishwavidyalaya Khairagarh (C.G.) India.

Abstract

When a coloured alkali halide crystal is exposed to a laser pulse, it will produce strain then the rate of interaction of F-centres with dislocations. In the expansion region of dislocations, the average energy E_1 of F-centre interacting with dislocations will lie between the normal ground state of F-centres and dislocation band. When the dislocation containing electrons are moving in a crystal, then the electrons may recombine with the defect centers containing holes also with the deep trap present in the crystals. The retrapping of dislocation electrons in the negative ion vacancies may also take place. The decay time will be controlled by pinning time of dislocations.

When a II-VI semiconductor exposed to laser pulse they will produce a shock-wave in the crystal and consequently the deformation of crystal takes place. Similarly we obtain the rate of generation of electron in the shallow traps and number of electrons in the shallow trap finally we obtain the maximum intensity and corresponding time. The decay time of ML will be equal to the life time of electrons in the shallow

traps. The ratio $\frac{I_{m_2}}{I_{m_1}}$ depend on the probability of transfer of

electrons from the conduction band to shallow traps and on the ratio of pinning time of dislocation and life time of electrons in the shallow traps.

Keywords: Luminescence, Mechano Luminescence (ML)

INTRODUCTION

Luminescence induced during mechanical deformation of solids is known as mechanoluminescence (ML). ML links mechanical spectroscopic, electrical structural and other properties of solids. A large number of organic and inorganic solids exhibits the phenomenon of ML can excited by grinding, rubbing, cutting cleaving, compressing or by impulsive deformation of solids [1,2]. It can be excited by the thermal shocks produced during sudden cooling or heating of the solids. Primarily ML can be classified into two types : deformation ML and tribo ML. Deformation ML is produced during the deformation of solids and tribo ML is produced during the rubbing of two materials or during the separation of two materials or in contact. On the basis of the deformation in solids needed for producing ML, deformation ML can further

be subdivided into three types namely, elastico ML, plastico ML and fracto ML. The ML occurring during elastico deformation of solids is called elastico ML, and the ML occurring during fracture of solids is called fractio ML.

We have been interested whether ML could be made to occur using a high energy laser pulses as the stress inducing agent [3]. The present chapter reports the theory of laser ML in coloured alkali halide crystals and II-VI semiconductors. In analogy with the laser thermoluminescence the ML caused by the deformation owing to laser pulses may be called as laser ML. The others materials like elemental and III-V semiconductors exhibit ML during their fracture, which is not related to the movement of dislocations, hence laser ML in coloured alkali halide crystals and this semiconductors may be observed during only when intense laser shocks pulses instead of direct deformation of solids and laser will create cracks in this semiconductor as such theory of laser ML in coloured alkali halide crystals and these semiconductors may be quite different from that of II-VI semiconductors whereby intense ML is observed during the direct deformation of solids and the movement of dislocations in these semiconductors.

LUMINESCENCE STIMULATED BY LASER SHOCKS IN COLOURED ALKALI HALID CRYSTALS

In a crystal having N_d dislocation of unit length per unit volume, if r_F is radius of interaction of dislocation with F-centres and v_d is the average velocity of dislocations, then in unit time, N_d dislocations may interact with the F-centres lying the volume $N_d v_d r_F$. If n_F is the density of F-centres, then for a crystal of unit volume, the rate of interaction of F-centres with dislocations may be given by

$$g_i = N_d v_d r_F n_F = \frac{\dot{\epsilon}}{b} r_F n_F \quad \text{----- (1)}$$

where b is the Burgers vectors and $\dot{\epsilon} = N_d v_d b$, is the strain rate.

Suppose a coloured alkali halide crystal is exposed to a laser pulse whose intensity is given by $I = I_0 \exp(-t / \tau_L)$, where I_0 is the maximum intensity and τ_L is the duration of a laser pulse. The laser pulse will produce strain in the crystal and the time dependence of strain rate $\dot{\epsilon}$ produced in the crystal may be given by

$$\dot{g} = AI_o \exp(-t / \tau_L) \quad \text{-----}(2)$$

where A is the correlating factor between g_i and intensity of laser pulse.

Thus from eqs(1) and (2), we get

$$g_i = \frac{AI_o \exp(-t / \tau_L) r_F n_F}{b}$$

or $g_i = g_o \exp(-t / \tau_L) \quad \text{-----}(3)$

where $g_o = \frac{AI_o r_F n_F}{b}$

In the expansion region of dislocations, the average energy E_i of F-centres interacting with dislocations is higher as compared to the non-interacting F-centres [1]. Thus, E_i will be lie between the normal ground state of F-centres and the dislocation band. If α_1 is the rate constant for jumping of interacting F-centres electrons to the dislocation band, and α_2 is the rate constant for the dropping back to the normal F-level, then we can write the following rate equation

$$\begin{aligned} \frac{dn_i}{dt} &= g_i - (\alpha_1 + \alpha_2)n_i \\ &= g_i - \alpha n_i \end{aligned} \quad \text{-----}(4)$$

where $\frac{1}{(\alpha_1 + \alpha_2)} = \tau_L$ is the lifetime of interacting F-centres and n_i is the number of F-centre electrons at any time t and $\alpha = (\alpha_1 + \alpha_2)$.

from eqs (3) and (4), we get

$$\frac{dn_i}{dt} = g_o \exp(-\alpha_L t) - \alpha n_i \quad \text{-----}(5)$$

where $\alpha_L = \frac{1}{\tau_L}$

eqs (5) may be written as

$$\frac{dn_i}{dt} + \alpha n_i = g_o e^{-\alpha_L t} \quad \text{-----}(6)$$

Integration of eq (6) for this integrating factor.

$$\begin{aligned} I.F. &= e^{\int \alpha \cdot dt} \\ &= e^{\alpha t} \end{aligned}$$

Thus the solution of eq (6) can be expressed as

$$n_i e^{\alpha t} = \int e^{\alpha t} \cdot g_o e^{-\alpha_L t} dt + C$$

where C is the integration constant

$$\begin{aligned} n_i e^{\alpha t} &= g_o \int e^{(\alpha - \alpha_L)t} dt + C \\ &= g_o \frac{e^{(\alpha - \alpha_L)t}}{(\alpha - \alpha_L)} + C \end{aligned} \quad \text{-----}(7)$$

For $n_i = 0$ at $t = 0$

$$C = \frac{-g_o}{(\alpha - \alpha_L)} \quad \text{-----}(8)$$

From eqs (7) and (8) we get

$$\begin{aligned} n_i e^{\alpha t} &= g_o \frac{e^{(\alpha - \alpha_L)t}}{(\alpha - \alpha_L)} - \frac{g_o}{(\alpha - \alpha_L)} \\ \text{or } n_i &= g_o \frac{e^{-\alpha_L t}}{(\alpha - \alpha_L)} - \frac{g_o e^{-\alpha t}}{(\alpha - \alpha_L)} \\ &= \frac{g_o}{(\alpha - \alpha_L)} \left[e^{-\alpha_L t} - e^{-\alpha t} \right] \\ &= -\frac{g_o}{(\alpha_L - \alpha)} \left[e^{-\alpha_L t} - e^{-\alpha t} \right] \end{aligned} \quad \text{-----}(9)$$

Using eq (9) the rate of generation of electrons in the dislocation band may be written as

$$g = \alpha_i n_i$$

or $g = -\frac{\alpha_1 g_o}{(\alpha_L - \alpha)} \left[e^{-\alpha_L t} - e^{-\alpha t} \right] \quad \text{-----}(10)$

for $\alpha_L \gg \alpha$ eq. (10) may be expressed as

$$g = \frac{\alpha_1 g_o}{\alpha_L} \cdot e^{-\alpha t} \quad \text{-----}(11)$$

When the dislocations containing electrons are moving in a crystal, then the electrons may recombine with the defect centers containing holes also with the deep trap present in the crystals. The retrapping of dislocation electrons in the negative ion vacancies may also take place. Suppose N_1, N_2 and N_3 are densities of recombination centers, deep traps and negative ion vacancies (without trapped electrons), respectively, and σ_1, σ_2 and σ_3 are the capture cross sections for the recombination centers, deep trap and negative ion vacancies, respectively, then the rate equation may be written as

$$\frac{dn_d}{dt} = g_o e^{-\alpha t} - \sigma_1 N_1 v_d n_d - \sigma_2 N_2 v_d n_d - \sigma_3 N_3 v_d n_d$$

or
$$\frac{dn_d}{dt} = g_o e^{-\alpha t} - n_d / \tau_d$$

or
$$\frac{dn_d}{dt} = g_o e^{-\alpha t} - \alpha_d n_d \quad \text{-----(12)}$$

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

and
$$\alpha_d = \frac{1}{\tau_d}$$

Equation (12) may be written as

$$\frac{dn_d}{dt} + \alpha_d n_d = g_o e^{-\alpha t} \quad \text{-----(13)}$$

Integration of eqs. (13) for this integrating factor

$$\begin{aligned} \text{I.F.} &= e^{\int \alpha_d t} \\ &= e^{\alpha_d t} \end{aligned}$$

Then the solution of equation (13) can be expressed as

$$\begin{aligned} n_d e^{\alpha_d t} &= \int e^{\alpha_d t} g_o e^{-\alpha t} dt + c \\ &= g_o \int e^{(\alpha_d - \alpha)t} dt + c \\ &= g_o \frac{e^{(\alpha_d - \alpha)t}}{(\alpha_d - \alpha)} + c \quad \text{-----(14)} \end{aligned}$$

For $n_d = 0$ at $t = 0$, then we get

$$C = -\frac{g_o}{(\alpha_d - \alpha)} \quad \text{-----(15)}$$

From eqs (14) and (15) we get

$$\begin{aligned} n_d e^{\alpha_d t} &= g_o \frac{e^{(\alpha_d - \alpha)t}}{(\alpha_d - \alpha)} - \frac{g_o}{(\alpha_d - \alpha)} \\ \text{or } n_d &= \frac{g_o}{(\alpha_d - \alpha)} \left[e^{-\alpha t} - e^{-\alpha_d t} \right] \quad \text{-----(16)} \end{aligned}$$

If η is the probability of radiative recombination, then the deformation induced ML intensity for a crystal of volume V containing Nd dislocations of unit length may be expressed as

$$\begin{aligned} I &= \eta \sigma_1 N_1 v_d n_d \\ \text{or } I &= \frac{\eta \sigma_1 N_1 v_d A I_o r_F n_F}{b(\alpha_d - \alpha)} \left[e^{-\alpha t} - e^{-\alpha_d t} \right] \quad \text{-----(17)} \end{aligned}$$

Equation (17) indicates that I_o should be maximum for a particular value of time t given by

$$t_m = \frac{1}{(\alpha_1 - \alpha)} \ln\left(\frac{\alpha_d}{\alpha}\right) = \frac{1}{(\alpha_L - \alpha)} \ln\left(\frac{\alpha_d}{\alpha}\right) \quad \text{-----(18)}$$

For $\alpha_d t \gg 1$, eq (17) may be expressed as

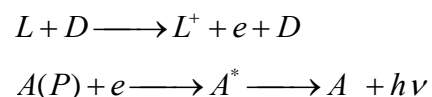
$$I = \frac{\eta \sigma_1 N_1 v_d A I_o r_F n_F}{b(\alpha_d - \alpha)} e^{-\alpha t} \quad \text{-----(19)}$$

Eqⁿ. (19) shows the exponential decay of ML intensity I, where the decay time will be controlled by α , i.e., the pinning time of dislocations.

It is to be noted that the electrons captured by dislocations have two types of motions, firstly, they move with dislocations, and secondly they also move along the dislocation axis with a very low velocity of the order of 0.1cm/sec. Thus, initially the ML intensity should decay with a fast rate and then it should decay with a slow rate. The first decay time should give the pinning time of dislocations and second lifetime of the electrons in the dislocation band.

LUMINESCENCE STIMULATED BY LASER-SHOCKS IN II-VI SEMICONDUCTORS

An analysis of the possible ML mechanisms has shown that the interacting between charged dislocations and activator centers leads to activator ionization. Ionization may occur by tunneling of electrons from the impurity level of the conduction band under the operation of strong electric field close to a charged dislocation [4-7]. The electric field at a distance r from the core of a charged dislocation is given by $E = 2q / \epsilon_o r$, where q is the linear charge density of dislocation, ϵ_o is the dielectric permittivity. At $q = 0.35$ e/units, where e is the electronic charge, the electric field at a distance $r = 10^{-7}$ cm from the dislocation core is $E = 3.4 \times 10^6$ volt/cm¹. In this case, the electric field due to charged dislocations ionizes the electrons from filled traps and the subsequent recombination of electrons with the activator centres containing holes gives rise to luminescence. The process can be represented as follows :



where L is a trap filled with electron, L^+ is an empty trap and A(P) is the activator containing hole.

Supposing a II-VI semiconductor like ZnS is exposed to an infrared laser pulse whose intensity is given by $I = I_o \exp(-\delta t)$, where I_o is the intensity at $t = 0$ and δ is a factor inversely related to the pulse duration of laser. It is known that the laser pulse produces a shock-wave in the crystal and consequently the deformation of crystal takes

place [8-10]. For a laser pulse of short duration and low pulse energy, the local heating will not be significant and consequently the intensity of black body radiation may be assumed to be negligible as compared to that of the laser ML produced in the bulk of crystal. It is to be noted II-VI semiconductors exhibit ML even during their plastic deformation.

Let us assume that the time dependence of the rate of generation g_d of moving dislocations in the localized region of the crystal due to the laser pulse is given by

$$g_d = BI_o \exp(-\delta t) \quad \text{-----}(20)$$

where B is correlating factor between g_d and the intensity of the laser pulse.

If r_i is the radius of interaction between moving dislocations and the filled traps in the crystal and λ is the mean distance traveled by a dislocation before its pinning, then the volume in which g_d dislocation of unit length can interact per unit time will be $2r_i\lambda g_d$.

If N_t is the concentration of filled traps, then the number of filled traps interacting with the dislocations per unit time is given by

$$g_i = 2r_i\lambda g_d N_t \quad \text{-----}(21)$$

From eqs. (20) and (21), we get

$$g_i = 2r_i\lambda N_t BI_o \exp(-\delta t) \quad \text{-----}(22)$$

If α_1 is the rate constant for tunneling of electrons from the filled traps to the conduction band and α_2 is the rate constant for the dropping back of the trapped electrons, then we may write the following rate equation –

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

or
$$\frac{dn_i}{dt} = g_i - \alpha n_i \quad \text{-----}(23)$$

where g_i is the number of interacting filled traps at any time t and $\tau_p = 1/(\alpha_1 + \alpha_2) = \frac{1}{\alpha}$ is the lifetime of interacting filled traps or the pinning time of dislocations (Hayashiuchi et al.).

From eqs. (22) and (23), we get

$$\frac{dn_i}{dt} = 2r_i\lambda N_t BI_o \exp(-\delta t) - \alpha n_i \quad \text{-----}(24)$$

Integrating eq. (24) and taking $n_i = 0$, at $t = 0$, we get

$$n_i = \frac{2r_i\lambda N_t BI_o}{(\delta - \alpha)} [\exp(-\alpha t) - \exp(-\delta t)] \quad \text{----}(25)$$

Thus the rate of generation of electrons in the conduction band may be given by

$$g = \alpha_i n_i = \frac{2\alpha_i r_i \lambda N_t BI_o}{(\delta - \alpha)} [\exp(-\alpha t) - \exp(-\delta t)] \quad \text{----}(26)$$

If β_1 and β_2 are the rate constants for the radiative and non-radiative recombination of electrons with the hole centers, respectively, and β_3 is the rate constants for the transfer of electrons to the shallow traps, then we may write the following rate equation

$$\frac{d(\Delta n)}{dt} = g - (\beta_1 + \beta_2 + \beta_3)\Delta n$$

or
$$\frac{d(\Delta n)}{dt} = \frac{2\alpha_i r_i \lambda N_t BI_o}{(\delta - \alpha)} [\exp(-\alpha t) - \exp(-\delta t)] - \beta \Delta n, \quad \text{---}(27)$$

where Δn is the change in the number of electrons in the conduction band at any time t, $\beta = (\beta_1 + \beta_2 + \beta_3)$ and $1/\beta = \tau$, is the lifetime of electrons in the conduction band.

Integrating eq. (27) and taking $\Delta n = 0$ at $t = 0$, we get

$$\Delta n = \frac{2\alpha_i r_i \lambda N_t BI_o}{(\delta - \alpha)} \left[\frac{\exp(-\alpha t)}{(\beta - \alpha)} - \frac{\exp(-\delta t)}{(\beta - \alpha)} + \frac{(\delta - \alpha)\exp(-\beta t)}{(\beta - \delta)(\beta - \alpha)} \right] \quad \text{---}(28)$$

For a laser-pulse of short duration, $\delta \gg \alpha$ and $\delta \gg \beta$, and in the deformation region $\beta > \alpha$. Thus, eq. (28) may be expressed as

$$\Delta n = \frac{2\alpha_i r_i \lambda N_t BI_o}{\delta(\beta - \alpha)} [\exp(-\alpha t) - \exp(-\beta t)] \quad \text{---}(29)$$

Thus, the intensity of transient ML may be given by

$$I_1 = \beta_1 \Delta n = \frac{2\beta_1 \alpha_i r_i \lambda N_t BI_o}{\delta(\beta - \alpha)} [\exp(-\alpha t) - \exp(-\beta t)] \quad \text{---}(30)$$

Equation (30) indicates that the ML intensity should attain a maximum value I_{m1} for a particular value of time t_{m1} . For $\beta > \alpha$ using equation (30) t_{m1} and I_{m1} may be given by

$$t_{m1} = \frac{1}{(\beta - \alpha)} \ln\left(\frac{\beta}{\alpha}\right) \quad \text{---}(31)$$

and

$$I_{m1} = \frac{2\alpha_i \beta_1 r_i \lambda N_t BI_o}{\delta \beta} \quad \text{---}(32)$$

As $\beta \gg \alpha$ for $\beta t \gg t$ eq. (11) be expressed as

$$I_1^d = \frac{2\alpha_i\beta_i r_i \lambda N_i B I_o}{\delta(\beta - \alpha)} \exp(t / \tau_p) \quad \text{-----(33)}$$

Equation (33) indicates the exponential decay of ML intensity ML, I_1 in which the decay time of ML will give the lifetime of interacting filled traps or the pinning time of dislocations.

For $\beta \gg 1$ equation (29) may be expressed as

$$\Delta n = \frac{2\alpha_i r_i \lambda N_i B I_o}{\delta(\beta - \alpha)} \exp(-\alpha t) \quad \text{-----(34)}$$

Thus, the rate of generation of electrons in shallow traps may be given by

$$G = \beta_3 \Delta n = \frac{2\alpha_i \beta_3 \lambda N B I_o}{\delta(\beta - \alpha)} \exp(-\alpha t) \quad \text{-----(35)}$$

If τ_s is the lifetime of electrons in the shallow traps, then we may write the

$$\frac{d\Delta n_s}{dt} = G - \left[\frac{n_s}{\tau_s} \right]$$

following rate equation

$$\text{or } \frac{d\Delta n_s}{dt} = \frac{2\alpha_i \beta_3 r_i \lambda N_t B I_o}{\delta(\beta - \alpha)} \exp(-\alpha t) - \gamma n_s \quad \text{-----(36)}$$

where Δn_s is the number of electrons in the shallow traps at

any time t , and $\tau_s = \frac{1}{\gamma}$. Integrating equation (36) and taking

$n_s = 0$, at $t = 0$, we get

$$\text{or } \Delta n_s = \frac{2\alpha_i \beta_3 r_i \lambda N_t B I_o}{\delta(\beta - \alpha)(\alpha - \gamma)} [\exp(-\gamma t) - \exp(-\alpha t)] \quad \text{-----(37)}$$

If η is the probability of radiative recombination of electrons released from the shallow traps, then the intensity I_2 of the delayed ML may be given by

$$I_2 = \eta \Delta n_s \gamma = \frac{2\eta \gamma \alpha_i \beta_3 r_i \lambda N_t B I_o}{\delta(\beta - \alpha)(\alpha - \gamma)} [\exp(-\gamma t) - \exp(-\alpha t)] \quad \text{-----(38)}$$

The above equation indicates that the intensity of delayed ML should attain a maximum value I_{m2} for a particular value of time t_{m2} . For $\alpha > \gamma$, using equation (38), t_{m2} and I_{m2} may be given by

$$t_{m2} = \frac{1}{(\alpha - \gamma)} \ln \left[\frac{\alpha}{\gamma} \right] \quad \text{-----(39)}$$

$$\text{and, } I_{m2} = \frac{2\eta \gamma \alpha_i \beta_3 r_i \lambda N_t B I_o}{\delta(\beta - \alpha)\alpha} \quad \text{-----(40)}$$

As $\alpha > \gamma$, for $\alpha t \gg 1$, from equation (38) the decay of ML intensity I_2 may be given by

$$I_2^d = \frac{2\eta \gamma \alpha_i \beta_3 r_i \lambda N_t B I_o}{\delta(\beta - \alpha)(\alpha - \gamma)} \exp\left(\frac{-\gamma t}{\tau_s}\right) \quad \text{-----(41)}$$

Equation (41) indicates the exponential decay of ML intensity I_2 , in which the decay time of ML will be equal to

the life time of electrons in the shallow-traps. As $\eta = \frac{\beta_1}{\beta}$

and $\beta \gg \alpha$, from eqs. (32) and (41), the ratio of I_{m2} and I_{m1} is given by

$$\frac{I_{m2}}{I_{m1}} = \frac{\beta_3 \gamma}{\beta \alpha} = P_t \frac{\tau_p}{\tau_s} \quad \text{-----(42)}$$

where $P_t = \frac{\beta_3}{\beta}$ is the probability of transfer of electrons

from the conduction band to shallow traps.

The above equation shows that the ratio I_{m2} / I_{m1} should depend on the probability of transfer of electrons from the conduction band to the shallow traps and on the ratio of pinning time of dislocation and the lifetime electrons in the shallow traps.

EXPERIMENTAL SUPPORT

Induce luminescence in several non-coloured organic and inorganic crystals by a 20 ns, 1060 nm pulse from a Nd glass laser whose pulse energy varied from 0.5 to 4 J cm⁻² ($\approx 200\text{MW}$ peak power). The spectra of laser induced emission were obtained by using a silicon intensified (SIT) vidicon detector and a multichannel analyser. So far as the time dependence of ML in II-VI semiconductors is concerned our preliminary observations have shown the occurrence of two ML peaks. This needs a further detailed investigation. So far as II-VI semiconductors are concerned no experimental study has been made to date.

CONCLUSIONS

The important conclusions drawn from the present investigation are as given below :

- (i) When the ML in a coloured alkali halide crystals or II-VI semiconductors is excited by the deformation caused by a laser pulse, then the ML intensity versus time curve should possess two peaks, where the first peak should occur in the region where deformation takes place owing to laser pulse and the second peak

should occur in the post-deformation region.

- (ii) In the laser-stimulated ML in coloured alkali halide crystals, the ML intensity should decay with a fast rate and then it should decay with a slow rate. The first decay time should give the pinning time of dislocations and second lifetime of the electrons in the dislocation band.
- (iii) In the core of II-VI semiconductors, the decay time of ML after t_{m1} should give the pinning time of dislocations and the decay time of ML after t_{m2} should give the lifetime of electrons in the shallow traps.
- (iv) In the core of II-VI semiconductors, the ratio I_{m2} / I_{m1} should be given by the product of probability of transfer of electrons from conduction band to shallow traps and the ratio of pinning time of dislocations and the lifetime of electrons in shallow traps.
- (v) By using laser pulse and an optical fiber, the ML may be observed owing to the movement of a single dislocation in coloured alkali halide crystals and II-VI semiconductors

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