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Ultrasonic pulse induced mechanoluminescence of europium doped strontium aluminate micro-crystals

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Abstract
When rare earth doped strontium aluminate micro-crystals are exposed to ultrasonic pulse, then mechanumnescence (ML) is induced. The intensity of ML is proportional to the power of ultrasonic pulse used for ML excitation. The ML in europium doped strontium aluminate micro-crystals can be understood on the basis of the piezoelectrically induced electron detrapping model. On the basis of the piezoelectrically-induced electron detrapping model, expressions are derived for the general kinetics of ML intensity, rise of ML intensity, peak ML intensity and decay of ML intensity, in which good agreement is found between the theoretical and experimental results. A linear relation between the ML intensity and the ultrasonic power can also be understood on the basis of the proposed theory. The present investigation shows that the ML can be used to detecting the presence of ultrasonic waves and also for the measurement of ultrasonic power.

Keywords: Ultrasonic pulse, mechanoluminescence, strontium aluminate

INTRODUCTION

Mechanoluminescence (ML) is a type of luminescence induced by any mechanical action on solids. The light emission induced by elastic deformation, plastic deformation and fracture of solids are called elastico ML (EML), plastico ML(PML) and fracto ML(FML), respectively [1]. ML can be induced by a new means, irradiation of ultrasonic pulse which produces intense luminescence. The intensity of ML is proportional to the power of irradiated ultrasonic pulse. [2-3].

The development of materials with strong ML intensity is an important goal in exploring application of ML in mechano-optical devices. In recent past, systematic materials research has been done and it has resulted in producing a variety of materials that emit an intensive and repeatable M without destruction. So far the most promising mechanoluminescent material is europium doped strontium aluminate [4]. For better understanding of mechanoluminesent materials and for exploring their applications in various purposes, significant theoretical studies are needed. The present paper reports the ultrasonic wave induced mechanoluminescence of europium doped strontium aluminate micro-crystals, and makes the comparison between the theoretical and experimental results.

Mechanism of the ML of Europium Doped Strontium Aluminate Micro-crystals

The appearance of ML only in the piezoelectric- phase of strontium aluminate phosphors indicates that the piezoelectrification is responsible for the ML of europium doped strontium aluminate micro-crystals [5]. The steps involved in the EML emission in europium doped strontium aluminate phosphors are as given below:
(i) The pressure produced by the ultrasonic pulse produces piezoelectric field in europium doped strontium aluminate micro-crystals because they are non-centrosymmetric, in which the piezoelectric field near certain defects centres may be high due to the change in the local structure.

I. The piezoelectric field reduces the trap-depth of the traps in the crystals
II. The decrease in trap-depth causes transfer of electrons from electron traps to the conduction band
III. Subsequently, the moving electrons in the conduction band are captured in the excited state of Eu^{2+} ions located at the bottom of the conduction band, whereby excited (Eu^{2+}) ions are produced.
IV. The de-excitation of $(Eu^{2+})^*$ ions gives rise to the light emission characteristic of the (Eu^{2+}) ions.

Theory

k be expressed as

$$P = P_o\left[1-\exp\left(-\frac{t}{\tau_r}\right)\right] \; P = P_o\left[1-\exp(-\xi t)\right] \qquad (1)$$

where $\xi = 1/\tau_r$, is the rate constant for the rise of pressure and P_o is the peak value of the pressure produced during exposure of the sample to ultrasonic pulse.

If Ω is the activation volume near an activator ion at which the piezoelectric constant is high, N_l is the number of activators in the sample and N_t is the concentration of traps in the sample, then the total number of traps in the sample is given by, $N_0 = \Omega N_l N_t$. Considering the exponential distribution of traps in the activation volume near the activator ions in the crystallites, the number of traps N (E_i) of energy E_i is given by the following Boltzmann statistical formula

$$N(E_i) = ZN_0 \exp(-ZE_i) \quad (2)$$

where N_0 is the total number of traps in the activation volume of the sample and Z=1/kT is the distribution coefficient, in which k is the Boltzmann constant and T is the absolute temperature of the phosphor material.

Up to the threshold energy E_{th} the trap-depths are comparable to kT, and therefore, the shallow traps lying between zero and E_{th} are thermally detrapped. Thus, the detrappable traps will be present only from E_{th} to E and using Eq. (2), the total number N_t of detrappable traps from E_{th} to E can be expressed as

$$N_t = \int_{E_{th}}^{E} ZN_0 \exp(-ZE)dE$$

$$N_t = N_0[\exp(-ZE_{th}) - \exp(-ZE)] \quad (3)$$

When an external pressure is applied, then the piezoelectric field F is produced. If α is the decrease in the trap-depth per unit electric field, then the decrease in trap-depth for the field F will be αF. Due to the decrease in trap-depth detrapping of electrons will take place and consequently the total number of filled traps will decrease. If N_d is the total number of detrapped traps after change in the trap-depth αF, then from Eq. (3), we get

$$N_d = N_0[\{\exp(-Z\alpha F_{th}) - \exp(-Z\alpha F)\}] \quad (4)$$

where F_{th} is the threshold piezoelectric field for the ML. Differentiating Eq. (4), we get

$$\frac{dN_d}{dF} = N_0 Z\alpha[\exp(-Z\alpha F)] \quad (5)$$

$$\text{or, } \frac{dN_d}{dt} = N_0 Z\alpha\dot{F}[\exp(-Z\alpha\dot{F}t)] \quad (6)$$

As the detrapped electrons are transferred to the conduction band, the rate of generation of electrons in the conduction band is given by

$$g = \frac{dN_d}{dt} = N_0 Z\alpha\dot{F}[\exp(-Z\alpha\dot{F}t)] \quad (7)$$

If B is the correlating factor between F and the piezoelectric charge Q, then F = BQ. For the piezoelectric constant d_0 near the localized piezoelectric region in the crystal, then for the applied pressure P and pressing rate $\dot{P}$, we get, $\dot{F} = B\dot{Q} = Bd_0\dot{P}$ and $F = BQ = Bd_0P$, and therefore, Eq. (7) can be expressed as

$$g = N_0 Z\alpha B d_0 \dot{P}[\exp(-Z\alpha\dot{F}t)] \quad (8)$$

From Eq.(1), $\frac{dP}{dt}$ can be expressed as $\frac{dP}{dt} = \dot{P} = P_o\xi\exp(-\xi t)$ (9)

Thus, from Eqs. (8) and (9), we get

$$g = N_0 Z\alpha B d_0 P_o \xi \exp(-\xi t)[\exp(-Z\alpha\dot{F}t)] \quad (10)$$

As $Z\alpha\dot{F}t$ is much less than 1 and ξ is a large quantity, Eq. (10) can be written as

$$g = N_0 Z\alpha B d_0 P_o \xi \exp(-\xi t) \quad (11)$$

If τ is the lifetime of electrons in the conduction band and v_d is the drift velocity of electrons, then the mean free path of the electrons is given by $\lambda = v_d\tau$. If μ is the mobility of electrons, then $v_d = \mu F$, and then, $\lambda = \mu F\tau = \mu d_0 P\tau$. Thus the rate of excitation of Eu^{2+} ions can be expressed as

$$R = N_0 Z\sigma n_r \lambda\alpha B d_0 P_o \xi \exp(-\xi t) \quad (12)$$

where σ capture cross-section of electrons and n_r is the concentration of Eu^{2+} centres, whose excited state capture the electrons moving in the conduction band.

Now, Eq. (12) can be written as

$$R = N_0 Z\sigma n_r \alpha B^2 d_0^2 P_0^2 \xi\mu\tau[\exp(-\xi t) - \exp(-2\xi t)] \quad (13)$$

If τ_e is the lifetime of the excited state of Eu^{2+} ions, then we can write the following rate equation

$$\frac{dN_{ex}}{dt} = N_0\sigma n_r \alpha B^2 d_0^2 P_0^2 \xi Z\mu\tau .$$
$$[\exp(-\xi t) - \exp(-2\xi t)] - \alpha N_{ex} \quad (14)$$

where $\alpha = 1/\tau_e$, and N_{ex} is the number of excited at any time t. Integrating Eq.(14) and taking $N_{ex} = 0$, at at t =0, we get,

$$N_{ex} = N_0\sigma n_r B^2 d_0^2 P_0^2 \xi Z\mu\tau .$$
$$[\frac{\exp(-\xi t)}{(\alpha-\xi)} - \frac{\exp(-2\xi t)}{(\alpha-2\xi)} + \frac{\xi\exp(-\alpha t)}{(\alpha-\xi)(\alpha-2\xi)}] \quad (15)$$

For **α>>,** Eq.(15) can be written as

$$N_{ex} = N_0\sigma n_r B^2 d_0^2 P_0^2 \xi Z\mu\tau[\exp(-\xi t) - \exp(-2\xi t)] \quad (16)$$

If η is the efficiency for the radiative decay of excited Eu^{2+} ions, then using Eq.(16) the ML intensity can be expressed as

$$I = \eta\alpha N_{ex} = \eta\alpha N_0\sigma n_r B^2 d_0^2 P_0^2 \xi Z\mu\tau .$$
$$[\exp(-\xi t) - \exp(-2\xi t)] \quad (17)$$

Substituting the value of $N_0 = \Omega N_l N_t$. Eq. (17) can be written as

$$I = \eta\alpha N_{ex} = \eta\alpha\Omega N_1 N_t\sigma n_r B^2 d_0^2 P_0^2 \xi\sigma n_r\mu\tau .$$
$$[\exp(-\xi t) - \exp(-2\xi t)] \quad (18)$$

Using Eq. (18), the rise of ML intensity I_r, peak value of ML intensity I_m and decay of ML intensity I_{df} can be expressed as

$$I_r = \eta\alpha\Omega N_1 N_t Z B^2 d_0^2 P_0^2 \xi^2\sigma n_r \mu\tau \quad (19)$$

$$I_m = \frac{\eta\alpha\Omega N_1 N_t Z B^2 d_0^2 P_0^2 \xi\sigma n_r \mu\tau}{4} \quad (20)$$

and, $I_{df} = \eta\alpha\Omega N_l N_t Z B^2 d_0^2 P_0^2 \xi\sigma n_r \mu\mu .$

$$\exp(-\xi t_m)\exp\{-\xi(t-t_m)\} \quad (21)$$

Experimental Support to the Proposed Theory

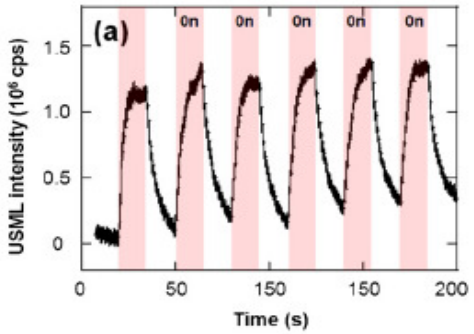

Fig 1. ML response of ultrasonic irradiation in europium doped strontium micro-cystals (after Terasaki et al. ref [2])

Fig. 1 shows ML response of ultrasonic irradiation [2]. It is seen that, as soon as the sample of europium doped strontium aluminate micro-crystals is exposed to ultrasonic pulse, initially the ML intensity increases with time, attains a peak value I_m at a particular time t_m, and later on it decreases with time t. As such, the kinetics of ML follows Eq. (18). It is seen from Fig. 1 that when the sample is exposed to ultrasonic pulse, initially the ML intensity increases linearly with time. This finding supports Eq. (19).

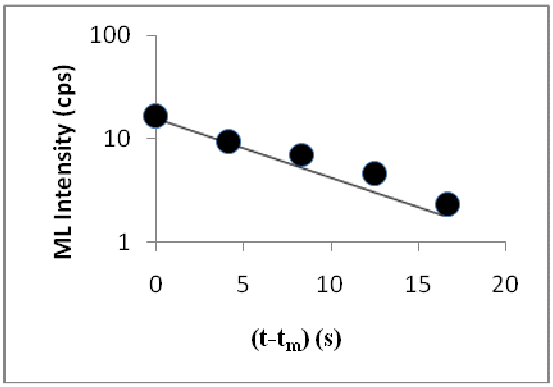

Fig 2. Semilog Plot of ML Intesity vs ($t-t_m$)

Fig. 2 shows the semilog plot of ML intensity versus ($t-t_m$). It is evident from Fig. 2 that the semilog plot of ML intensity I and ($t-t_m$) is a straight line with a negative slope. This result indicates the exponential decay of the ML intensity after t_m. Thus, the decay of ML intensity follows Eq. (21). Using Eq. (21) and Fig.2, the value of decay time of ML is found to be 8.47 s.

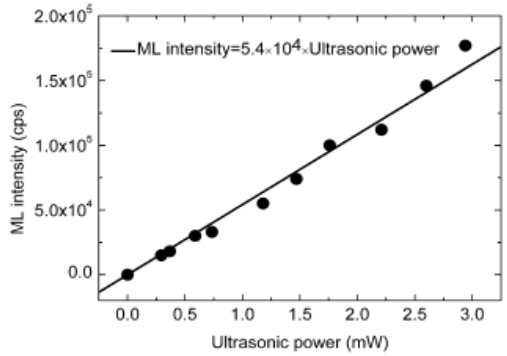

Fig 3. Influence of the ultrasonic power on the ML intensity (after Zhan et al. ref. [3])

Fig. 3 shows the relationship between the ML intensity and ultrasonic power [3]. As workdone will be proportional to P^2, the magnitude of power will be proportional to P_0^2. Hence, Eq. (19) indicates that the ML intensity should be proportional to the ultrasonic power. Thus, it seems that there is a good agreement between the theoretical and experimental results

CONCLUSIONS

Following the piezoelectrically-induced electron detrapping model of ML in europium doped strontium aluminate micro-crystal expressions are derived for the general kinetics of ML intensity I, rise of ML intensity I_r, peak ML intensity I_m and decay of ML intensity I_{df}, in which good agreement is found between the theoretical and experimental results. Moreover a linear relation between the ML intensity and the ultrasonic power can be understood on the basis of the proposed theory. It is shown that the time constant τ_r for the rise of ultrasonic induced pressure can be determined using mechanoluminescence. The present investigation shows that the ML can be used to detecting the presence of ultrasonic waves and also for the measurement of ultrasonic power.

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