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D.R. Mishra ^{a,}*, A.S. Bishnoi ^b, Anuj Soni ^a, N.S. Rawat ^a, B.C. Bhatt ^{a,1}, M.S. Kulkarni ^a, D.A.R. Babu ^a

^a Radiological Physics and Advisory Division, Bhabha Atomic Research Centre, Mumbai 400 085, India ^b Cdr. Indian Navy, IHQ-MOD(N)/DGNAI Office, West Block-V, Wing No-01, First Floor, R K Puram, New Delhi 110066, India

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ABSTRACT

The detailed theoretical and experimental approach to novel technique of pulse frequency modulated stimulation (PFMS) method has been described for NL-OSL phenomenon. This method involved pulsed frequency modulation with respect to time for fixed pulse width of 532 nm continuous wave (CW)-laser light. The linearly modulated (LM)-, non-linearly (NL)-stimulation profiles have been generated using fast electromagnetic optical shutter. The PFMS parameters have been determined for present experimental setup. The PFMS based LM-, NL-OSL studies have been carried out on dosimetry grade single crystal α -Al₂O₃:C. The photo ionization cross section of α -Al₂O₃:C has been found to be \sim 9.97 \times 10⁻¹⁹ cm² for 532 nm laser light using PFMS LM-OSL studies under assumption of first order of kinetic. This method of PFMS is found to be a potential alternative to generate different stimulation profiles using CW-light sources.

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1. Introduction

In recent years optically stimulated luminescence (OSL) has emerged as the novel technique of radiation measurements with widespread applications, such as archaeological/geological dating, personnel, environmental monitoring, medical dosimetry and more advanced space dosimetry for astronauts [\[1–4\].](#page--1-0) The OSL technique involves optical stimulating the meta-stable trapped charge carriers from wide band gap insulators/semiconductors by using mostly visible spectrum of light source and subsequent radiative recombination of these optically stimulated released charge carriers with polarity of opposite charges. Under condition that such material are previously exposed to ionizing radiation or UV light. In first case the electrons/holes generated due to band ionization and second case simply because of impurity defects ionization due to UV irradiation. The time integration of this emitted OSL signal is proportional to quantity of the radiation absorbed by the material. There are various modes of optical stimulation with generic names like, continuos wave (CW)-OSL, linearly modulated (LM)-OSL, non-linearly modulated (NL)-OSL) and pulsed (P)-OSL. In CW-OSL mode the stimulation intensity is held constant with respect to time, thus results in decay of OSL intensity with respect to time. In linearly modulated (LM) stimulation mode, the stimulation intensity increases linearly as a function of time over a sample results in OSL signal consists of multiple peaks (in intensity vs time plot) originating from different OSL charge traps defects having different values of photo-ionization cross-sections (PIC) [\[5\]](#page--1-0). The more generalized method of generating NL-stimulation intensity profiles has been suggested by Mishra et al. [6-7], which results in NL-OSL phenomenon. Under this method, stimulation intensity varies continuously and non-linearly with respect to time, results in OSL intensity curves may contains multiple peaks having better separation/resolution of very closely overlapping OSL components as compare to other conventional CW/LM mode of stimulation along with improved signal to noise (S/N) ratio. In the POSL mode, optical stimulation carried out using a series of short light pulses $($ <1 μ s) and emitted luminescence signal is recorded in between the stimulating pluses $[8]$. Such series of stimulation pulses are synchronized with gating of PMT output during stimulation periods, thus providing considerable improvement in S/N ratio. The POSL stimulation parameters have been greatly optimized for commercially available dosimetric grade α -Al₂O₃:C phosphor [\[8\]](#page--1-0), but for other OSL phosphors optimization of POSL parameters require, which will depending on PIC of OSL traps and luminescence recombination lifetime. Therefore, POSL technique remains specific as it requires exclusively short duration of laser/LED stimulation pulses with variable repetition rates up to few Hz to \sim 10 kHz (for α -Al₂O₃:C) in order to achieve optimum signal to noise ratio. Generating desirable pulse width of $\leq \mu s$ with variable output laser power is more difficult, as it requires laser in Q-switching mode, which is expensive and has limitation on pulse repetition rates. However, recent advancement in semiconductor light source, such

[⇑] Corresponding author. Tel.: +91 22 25590599.

E-mail address: devesh22@yahoo.com (D.R. Mishra).

¹ DST fellow.

as LED/semiconductor laser has opened new window of applications. Specifically LEDs as light source, which are nearly monochromatic and shortest period of light output pulse is limited by radiative luminescence lifetime (<ns) of charge recombination process. Depending upon the suitability with different stimulation OSL techniques various types of optical stimulation sources are in practice for OSL readout. For CW-stimulation, laser light, LEDs, or the blackbody radiation (incandescence) of tungsten halogen lamp can be used with appropriate combination of band/edge pass optical filters. However, generation of LM/NL-stimulations profiles needs good control on associated instrumentation more specifically the principle of light generation mechanism (which is light source specific) of these light sources. For the LEDs these stimulation profiles can be generated easily by controlling the LED forward bias current with appropriate close loop optical feedback control Mishra et al., 2011 [\[7,9\]](#page--1-0). But the generation of LM-stimulation profile show considerably sub-linearity in LED forward bias current vs optical output. This observed sub linearity can be attributed to heating of PN-junction due to high forward current which leads to loss of luminescence efficiency [\[9\].](#page--1-0) Therefore, some researchers have used pulse modulation of LED forward bias current to overcome from such self heating sub-linearity of LED current vs optical power [\[9\]](#page--1-0). Where, the duty cycle of LED forward bias current is varied linearly, thus producing linear increases in average optical stimulation power delivered at sample position. However, the effect of such averaging stimulation intensity on OSL phenomenon has not been explored completely either by theoretically or experimentally means. Achieving LM/NL stimulation profiles using laser has added advantage of mono-chromaticity and large dynamic stimulation power range from few W to hundreds of watt which is desired criteria for NL-OSL $[10]$ to get better resolution in closely overlapping OSL components. However, generation of such NL/LM-stimulation intensity using laser source required continuous varying laser power with respect to time in accordance with NL or LM stimulation profiles. This is difficult task as it requires appropriate reproducibility of optical power stabilizations associated with laser source design. Similar problem may occur in controlling desirable output optical power of conventional halogen or arc lamp specifically due to their slow time response and poor efficiency to generate optical intensity. On contrast, in POSL measurement pulsed laser has been used due to its high peak power, short duration and narrow energy bandwidth. The suitable synchronization of gating PMT output with laser pulse lead to highest signal to noise (S/N) ratio among all optical stimulation techniques. Apart from this, laser source offers great advantage of monochromatic light this helps in accurately determining the values of PIC and order of kinetics associated with charge transfer process in OSL phenomenon. In order to achieve LM/NL intensity from CW laser an amplitude modulation techniques can be adopted using opto-electric effects based devices [\[11,12\]](#page--1-0) and Pockels effect based modulators. In this the phase change occurs for polarizes light passing through in certain uniaxial crystal materials (such as, ADP – ammonium dihydrogen phosphate, KDP – potassium ammonium dihydrogen) [\[13\]](#page--1-0) which are under the stress of an electric field. This effect is a linear function of the voltage applied parallel to the crystal optical axis in the same direction as the incident light. Such modulators are expensive and need high voltage pulse ($\scriptstyle\sim$ 1–2 keV) to give the Pockels effects along with considerable loss in intensity of transmitted light beam. Apart from this, stimulation technique like POSL requires independent flexibilities in stimulation pulse width and pulse separation (frequency)/repetition rates. The possibility of modulation of CW-laser beam for LM/NL intensity will open a set of new potential to accurately measure the PIC associated with OSL active traps under monochromatic light. This will results in better estimation of order of kinetics asso-

ciated with OSL charge transfer phenomenon using fundamental

OSL rate equations. The present paper describes a detailed method to NL modulated CW-laser beam of 532 nm wavelength. This method utilizes the frequency modulation of fast electromagnetic solenoid shutter with associated optical setup designed get desirable CW-laser beam modulation. This modulated laser beam subsequently guided through optical fiber at the sample position and photodiode is used to measure the optical stimulation power at sample position. The LM-OSL and NL-OSL have been recorded by using this developed laser setup. The LM-, NL-, and pulsed-OSL stimulation intensity profiles have been generated and verified by photo diode. The OSL measurements of α -Al₂O₃:C phosphor has been carried out using these frequency modulated optical stimulation profiles.

2. Theory of pulse frequency modulated stimulation (PFMS)- OSL

2.1. Theory of method

In LM-OSL $[5]$ or NL-OSL $[6,7]$ methods, the OSL intensity is found to increase proportionally with stimulation intensity and reaches a maxima with respect to time, then subsequently decreases non-linearly to zero with further increase in stimulation intensity in respective cases. The time at which maxima in OSL intensity occurs dependent on $\text{PIC}(\sigma)$ of trap defects and the variation of stimulating intensity ramp rate γ (photons/cm²/s² or mW/cm²/s) in case of LM-stimulation or γ' (expressed in terms of photons fluence rate, i.e. photons/cm²/s^{*l*} where, *l* is a dimensionless parameter termed as time base power (TBP) of light intensity modulation) [\[5–7\].](#page--1-0) These OSL traps with different PIC values results in multiple peaks in LM/NL-OSL curve. One can assume quasi-equilibrium condition, that the rate of change of electron population in conduction band is negligibly small as compared to the rate of change of electrons concentration in metastable state under optical stimulation i.e.

$$
\frac{dn_c}{dt} \ll \frac{dn}{dt}, \frac{dm}{dt} \text{ and } n_c \ll n, m
$$

where, n_c and n are the concentrations of electrons in conduction band and OSL active traps respectively, m is the concentrations of holes in recombination trap centers. Thus, OSL intensity, I_{OSL} is proportional to rate of depletion of OSL active electron traps, i.e.

$$
I_{\text{OSL}} = -\frac{dn}{dt} \tag{1}
$$

In order to generalize PFMS theory for mathematical simplification, we assume here one-trap/one-recombination center model. So let us consider now electrons of concentration n are trapped at a localized states until stimulated into the conduction band by absorption of stimulation photons (of energy hv_{ex}). These stimulated released electrons are then able to recombine radiatively at trapped hole centers (thus emitting photons hv_{em} due to recombination of carriers of opposite nature) of concentrations m via conduction band, producing OSL with intensity $I_{\rm OSL}$. If ' f is optical excitation rate (s⁻¹) of optically released charges, under no re-trapping of optically released charge carriers (i.e. first-order kinetic), Eq. (1) can be re-written as OSL intensity

$$
I_{\text{OSL}} = -\frac{dm}{dt} = -\frac{dn}{dt} = nf \tag{2}
$$

where, f is product of photo-ionization cross-section $\sigma(\lambda)$ (cm²) associated with trap level and stimulating light flux $\phi(\lambda)$ (No. of photons/cm²/s or mW/cm²) can be expressed as

$$
f = \sigma(\lambda)\phi(\lambda) \tag{3a}
$$

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