

Continuous wave OSL in bulk AlN single crystals

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The kinetics of recombination luminescence of β -irradiated AlN single crystals has been studied with continuous wave optically stimulated luminescence (CW-OSL) method. It is shown that the OSL process is characterized by two exponential components with a decay time $\tau = 32$ and 212 s. Photoionization cross-sections $\sigma = 4.8 \cdot 10^{-19}$ and $7.3 \cdot 10^{-20}$ cm² have been evaluated, assum-

ing each component is related to electron traps based on V_N^- and/or O_N^- -centers at various charged states. It is established that dose dependences of the OSL response of the bulk crystals under study are linear with taking into correct accounting for the radiation induced afterglow and hence can be used for quantitative estimates in solid state dosimetry.

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1 Introduction Single crystalline AlN is a semiconductor with the band gap about 6.2 eV. It is characterised by high temperature conductivity, its thermal expansion coefficient being close to that of silicon, thus ensuring its wide use in optoelectronics and microelectronics as UV emitter and UV detectors, substrate for hybrid integrated chip and power electronics [1–3]. Synthesis of first bulk AlN single crystals gave the fillip to a number of studies related to investigation of their emission properties with different spectroscopic methods [4–11]. Possible applications of the AlN single crystals in solid state dosimetry of β - and UV-irradiation with methods of thermally and optically stimulated luminescence (TL and OSL) are shown in Ref. [12, 13]. It is known that intrinsic and impurity defects of the anion sublattice (V_N^- and O_N^- -centers) determine emission activity of AlN in the UV/Vis spectral range and help to detect and store dosimetric information [12–15]. Recently within the framework of formal kinetics analysis of the TL processes involving the above mentioned centers the values of the activation energy and effective frequency factor have been obtained [10, 13]; and dose dependences of the TL and OSL response have been analyzed [16, 17]. However, the basic parameters responsible for mechanisms of optically stimulated emission of oxygen-related complexes in bulk aluminum nitride have not been studied enough. In this context the present investigation was aimed at the study of kinetics of recombina-

tion luminescence and quantitative estimation of microparameters of capture centers in β -irradiated AlN single crystals using continuous wave optically stimulated luminescence (CW-OSL).

2 Samples and methods Single crystalline samples of aluminum nitride (Nitride Crystals, Inc.) were obtained from industrial fine-dispersed AlN powder of semiconducting purity preliminary annealed by the sublimation–recondensation method in high vacuum at 1900–2000 °C in nitrogen atmosphere [18]. Single crystalline SiC (0001) substrates were used to synthesize the samples with the growth rate 40–55 $\mu\text{m}\cdot\text{h}^{-1}$. According to the analysis data obtained by mass spectrometry with gas-discharge ionization, oxygen content in AlN does not exceed 10^{18} cm⁻³ [19]. The final samples had the form of disks 15 mm in diameter and 0.25–0.50 mm thick with epi-ready surfaces.

The CW-OSL and TL parameters were studied with custom-made spectrometric installation. The samples were irradiated by ⁹⁰Sr/⁹⁰Y β -source with the dose rate of 32 mGy·min⁻¹. Green DPSS laser (150 mW, 532 nm) was used for OSL measurements. Stimulating radiation intensity at the sample location was 25 mW·cm⁻². The data were registered in the 3.25 eV band with FWHM = 0.27 eV using a set of optical filters and a FEU-39A photomultiplier in counting regime.

The samples were pre-irradiated with dose D within the 2.7–960 mGy range. Then the AlN crystal was stored at room temperature for 5 min in darkness. Further the CW-OSL signal was registered for 10 min and the sample was annealed up to 700 K. This thermal treatment is required for complete emptying of charge carrier traps responsible for luminescence within the registered spectral range. Then the measurements were repeated for different irradiation dose. For $D = 64$ mGy the TL curves were registered before and after the CW-OSL measurement at heating rate $2 \text{ K}\cdot\text{s}^{-1}$.

3 Results Figure 1 shows the CW-OSL curves with varying dose of β -irradiation. It is seen that with increasing D the initial intensity and lightsum increase within the dose range under study.

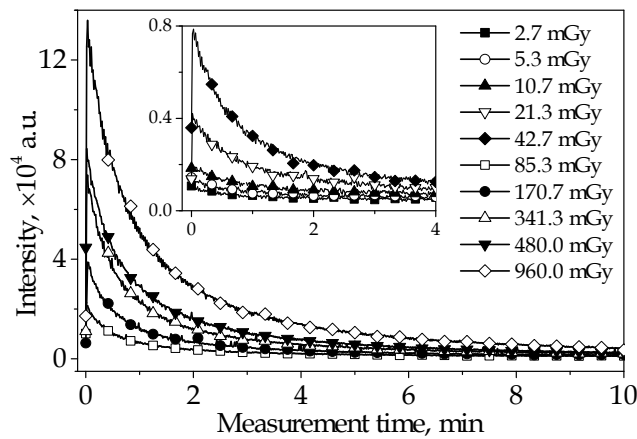


Figure 1 Experimental CW-OSL curves for β -irradiated AlN single crystal. Corresponding doses are shown in the figure.

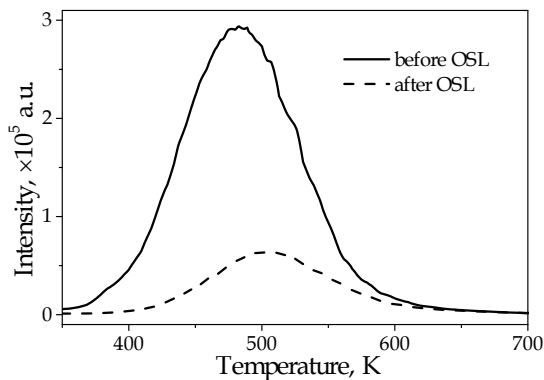


Figure 2 The TL curves for β -irradiated AlN single crystal.

Figure 2 demonstrates the measurement results of the TL response before and after OSL registration. Table 1 contains shape parameters of the TL peaks. It is seen that the TL curves look like a single wide unstructured peak, with maximum temperature (T_{max}), intensity (I_{max}) and lightsum (S_{TL}) varied under optical stimulation. However, halfwidth (ω) and form-factor (μ_g) are practically un-

changed. The value $\mu_g = 0.50$ corresponds neither to the processes of the first- nor of the second order kinetics, thus pointing to complex behavior of the registered thermally activated glow [20].

Table 1 Shape parameters of TL.

| parameter | $T_{max},$ $\pm 3 \text{ K}$ | $\omega,$ $\pm 3 \text{ K}$ | μ_g ± 0.01 | $I_{max},$ $\times 10^3 \text{ a.u.}$ | $S_{TL},$ $\times 10^6 \text{ a.u.}$ |
|------------|---------------------------------|--------------------------------|-----------------------|--|---|
| before OSL | 483 | 101 | 0.50 | 29.4 | 33.0 |
| after OSL | 506 | 104 | 0.50 | 6.4 | 7.6 |

4 Discussion With this measurement technique the CW-OSL signal is overlapped by the sample afterglow (AG) at room temperature after β -irradiation (dashed line in Fig. 3). Note, that AG is typical for different structures of the irradiated AlN and is present already before switching on the laser [11, 17]. After initiation of stimulation (“laser on”) the emission intensity increases dramatically and cumulative “OSL + AG” signal is registered (Fig. 3).

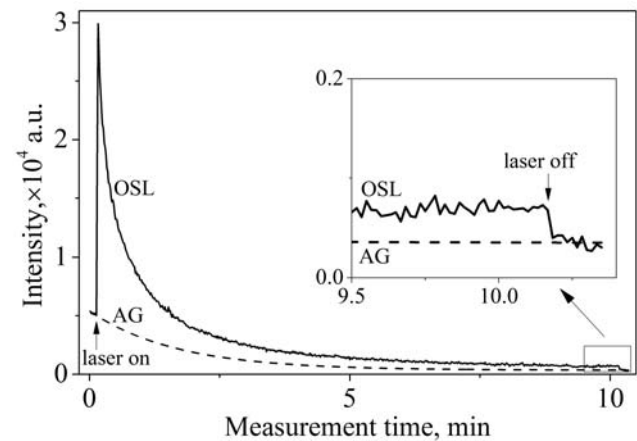


Figure 3 The CW-OSL (solid line) and AG (dashed line) curves for β -irradiated AlN single crystal with $D = 128$ mGy.

To analyze the CW-OSL kinetic features the effective signal was separated from the experimental curves (Fig. 1) by subtracting the AG curves. The latter were approximated by decay exponent. This mathematical approach is justified in Ref. [11], where AG of the irradiated AlN single crystals is shown to be described by exponential components with decay constants 59 s and 606 s. Thus, 5 min after the end of irradiation (before laser stimulation) basic contribution to AG is made by a slow component.

Kinetic analysis of the experimental CW-OSL curves including AG was done in terms of the first order formalism [21]:

$$I(t) = \sum_i I_{0i} \exp\left(-\frac{t}{\tau_i}\right), \quad (1)$$

where I_{0i} is the intensity at the initial moment of stimulation, a.u.; τ_i is the decay constant, s; t is the measurement time (or optical stimulation), s; i is the component number.

Figure 4 shows approximation example of the OSL curve by Eq. (1) for $i = 2$. Table 2 contains the parameter values of the CW-OSL curves calculated and R^2 determination coefficient. It is seen that all the OSL dependences are satisfactorily described by exponential components C1 and C2. This phenomenon can be explained by the presence of a number of optically active traps of charge carriers after β -irradiation, which are emptied at room temperature.

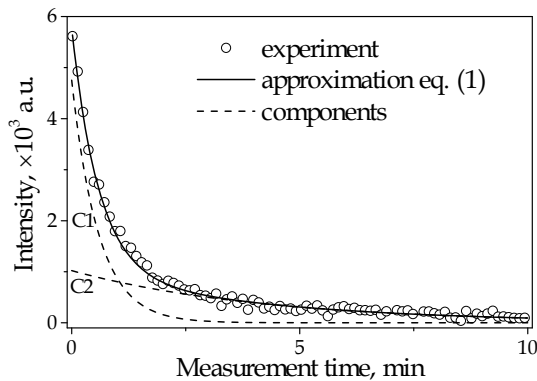


Figure 4 Example of the CW-OSL curve approximation by Eq. (1) for β -irradiated AlN single crystal with $D = 42.7$ mGy.

Table 2 Kinetic parameters of CW-OSL depending on the dose.

| D , mGy | $\tau \pm 0.5$, s | | σ , 10^{-19} cm ² | | R^2 |
|-----------|--------------------|-------|---------------------------------------|-----|-------|
| | C1 | C2 | C1 | C2 | |
| 2.7 | 19.4 | 30.3 | 7.7 | 4.9 | 0.871 |
| 5.3 | 10.9 | 79.3 | 13.6 | 1.9 | 0.930 |
| 10.7 | 30.9 | 146.3 | 4.8 | 1.0 | 0.960 |
| 21.3 | 19.1 | 86.9 | 7.8 | 1.7 | 0.988 |
| 42.7 | 35.3 | 250.3 | 4.2 | 6.0 | 0.994 |
| 85.3 | 23.6 | 120.1 | 6.3 | 1.2 | 0.997 |
| 170.7 | 28.5 | 208.5 | 5.2 | 0.7 | 0.996 |
| 341.3 | 31.7 | 236.6 | 4.7 | 0.6 | 0.997 |
| 480.0 | 33.1 | 223.7 | 4.5 | 0.7 | 0.998 |
| 960.0 | 37.5 | 232.7 | 4.0 | 0.6 | 0.998 |

Initial intensity I_0 of the OSL components increases with increasing irradiation dose within the range under study. This is in good agreement with the functional dependence $I_0 = n_0/\tau$, where n_0 is the concentration of the occupied traps for charge carriers directly before optical stimulation [21]. Obviously, concentration n_0 must grow with increasing radiation dose. Note that upon variation of D within the 42.7–960.0 mGy range the decay constants change slightly: $\tau = 32 \pm 5$ and 212 ± 43 s for C1 and C2, respectively.

Assuming each decay component of the OSL response to be related to an independent trap, let us estimate photoionization cross-sections of the capture centers with Eq. (2) [21]:

$$\sigma = \frac{1}{\tau\Phi}, \quad (2)$$

where $\Phi = 6.7 \times 10^{16}$ cm⁻²·s⁻¹ is stimulating radiation intensity.

Table 2 contains the obtained σ values of the traps responsible for C1 and C2. For $D = 42.7$ –960.0 mGy the above photoionization cross-sections change slightly: $\sigma = (4.8 \pm 0.7) \cdot 10^{-19}$ and $(7.3 \pm 1.5) \times 10^{-20}$ cm² for C1 and C2, respectively.

Considering the shape parameters of the TL curves (Table 1) it can be concluded that, optical depletion of the charge carrier traps responsible for the forming low-temperature part of peak dominates under laser stimulation of the irradiated sample. This is supported by the decrease of the lightsum and maximum intensity by 4.3 and 4.6, respectively as well as by the shift of the maximum toward the high-temperature range by 23 K. It is known that during storage of the AlN sample for 15 min in darkness the TL light sum decreases by 1.2 and the peak shifts by 5 K [11]. Similar changes of the TL peaks within the spectral range under study during optical stimulation and storage evidence that the same charge carrier traps take part in recombination OSL and AG processes of the irradiated bulk AlN single crystals. According to the analysis of independent experimental data one can assume that the OSL signal is caused at least by two types of traps which are thermally active within the RT–600 K range. They may be the anion V_N - and O_N -centers in different charged states, which form systems of allowed energy levels for electron capture in the irradiated AlN near the bottom of the conduction band.

Time measurements of the CW-OSL do not allow to assess the energy of defects levels in the forbidden band. However, it is known that the O_N -centers in different charge states form shallow electron traps [22], which are activated below RT and serve donors in the bleaching under study. In turn, the anion V_N -vacancies are deep donors with 0.5–0.9 eV estimates range of activation energy [10, 13]. In this regard, it can be assumed that the observed decay components C1 and C2 characterize optically stimulated ionization of the donor O_N - and V_N -centers, respectively.

The present study analyzes also the dose dependences of the light sum (S) and maximum intensity (I) of the OSL signal with and without taking into account the AG curves (Fig. 5). It is seen that the $S(D)$ curve with AG accounting is linear, the tangent slope being in two logarithmic coordinates $k = 1.02 \pm 0.01$. The $S(D)$ curve without AG is essentially nonlinear: $k = 0.87 \pm 0.01$. At the same time $k = 0.97 \pm 0.03$ for the $I(D)$ curve in both cases. Thus the OSL signal from β -irradiated AlN single crystals under study can be used to evaluate the absorbed dose.

AG affects essentially the CW-OSL dose dependences of the AlN powder and ceramics under UV-irradiation and the nonlinear dose dependence is assigned to the fading during irradiation [16, 17]. Note that the observed afterglow is one of the channels for fading of dosimetric infor-

mation. In this context the above data point to importance of accounting for AG when using the AlN single crystals as material for β -dosimetry with OSL method.

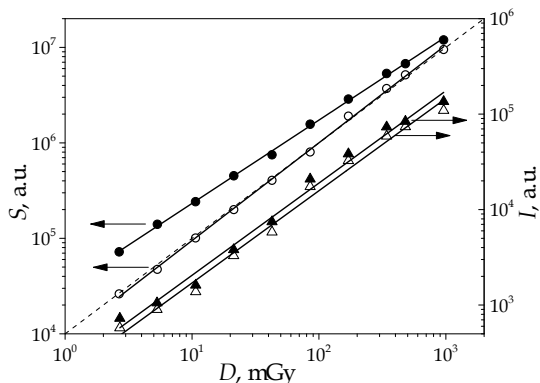


Figure 5 CW-OSL dose dependences for β -irradiated AlN single crystal with (open symbols) and without (solid symbols) taking into account the AG calculation. Solid lines – linear approximations; dashed line – ideal linear dependence ($k = 1$).

5 Conclusion Kinetic analysis of the OSL response of the irradiated AlN single crystals has been done within the framework of the first order kinetics formalism. It is shown that the luminescence process registered under optical stimulation of the samples after irradiation is characterized by fast and slow exponential components with the decay times $\tau = 32$ and 212 s, respectively. Photoionization cross-section of charge carrier traps $\sigma = 4.8 \times 10^{-19}$ and 7.3×10^{-20} cm², responsible for C1 and C2 components, respectively, in the OSL signal, was evaluated. On the basis of the experimental data and analysis of independent data it is shown that various charged states of the V_N^- - and O_N^- -centers may serve as electron traps participating in the recombination processes under study during optical or thermal stimulation of irradiated AlN single crystals. Optically active centers of capture are also responsible for the low-temperature part of the TL peak at 480 K. It is established that dose dependence of the OSL response in β -irradiated bulk AlN single crystals is linear with correct accounting for induced afterglow. Thus, the crystals under study can be used to detect β -irradiation and to solve the applied problems of OSL dosimetry.

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