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Peculiarities of Radiation Effects in MgO:Mn^{2+} Crystals

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Abstract

The thermally stimulated luminescence and IR absorption spectra of MgO:Mn^{2+} crystals irradiated in different reactors have been studied. It has been shown that dependence of TSL peaks (450 and 550 K) and optical absorption bands (3290, 3720 and 1600 cm^{-1}) on neutron fluence correlate. The trend of these changes in the same fluence range (10^{14} - 10^{17} n/cm^2) depends both on the initial state of the crystal and on the irradiation source, which may be explained by the "small dose effect".

Keywords: Magnesium Oxide, Radiation Defects, Thermally Stimulated Luminescence, IR Spectra

1. Introduction

In nominally pure (n.p.) single crystals of magnesium oxide steady increase of the thermally stimulated luminescence (TSL) peak intensity with growth of the reactor irradiation fluence has been generally observed [1,2]. In a number of cases for doped crystals, there exists a deviation from the rule. For instance, after irradiation of MgO:Mn^{2+} crystals by fast neutrons (4.5 MeV) in a fluence interval of 10^9 - 10^{11} n/cm^2 monotonic decrease of TSL intensity is observed [3], while at higher fluencies (>10^{13} n/cm^2) for the same crystals TSL increase takes place [2].

Analysis of the abovementioned results [1-3] makes it possible to suppose that differing behavior of TSL peaks is caused not by the difference in the ranges of neutron fluence, but is due to the differences in the irradiation sources and initial state of the crystals.

The presented paper is dealing with the detection and investigation of such and similar peculiarities of radiation effects in MgO:Mn^{2+} crystals.

2. Samples and Method

TSL curves were registered in a temperature interval of 300 - 775 K with a constant sample heating rate of 1 K/s [2].

The IR absorption spectra were measured in a wavelength interval of 4000 - 400 cm^{-1} at room temperature.

The samples irradiation was carried out by mixed (n,γ)-radiation in the channels of two different reactors:

1) IBR-2 (Dubna, Russia), where the differential fluxes of thermal and fast neutrons were equal to 4.1 × 10^{11} and 4.3 × 10^{11} n/cm^2·s respectively, γ-background was close to 300 krad/h;

2) WWR-SM (Tashkent, Uzbekistan), where the differential flux of thermal neutrons was 2.5 × 10^{13} n/cm^2·s, and the flux of fast neutrons was one third of that value.

Single crystalline samples of magnesium oxide (10 × 10 × 0.5 mm^3) were cleaved out from monocrystalline blocks (MTI Corporation). The impurities content in these samples, determined by EPR and IR spectroscopy, was 285 ppm of Mn^{2+} and 3.8 × 10^{17} cm^{-3} of OH^- respectively. According to the certificate of the MTI Corporation, the samples contained as well the following impurities: Ca < 40, Al < 15, Si < 10, Fe < 50, Cr < 10, B < 5, C < 10 ppm.

3. Results

3.1. Irradiation of MgO:Mn^{2+} Crystals in the Reactors WWR-SM and IBR-2

TSL curves of MgO:Mn^{2+} crystals irradiated in the reactor WWR-SM are given in **Figure 1**. The maximal intensity of the luminescence peak at 550 K (curve 1) is observed at the starting stage of irradiation (2 × 10^{14} n/cm^2); with the fluence increase the peak intensity decreases steadily (curves 2 - 5). The intensity of the peak at 450 K also drops after minor initial increase when changing the fluence from 2 × 10^{14} n/cm^2 to 1 × 10^{16} n/cm^2.

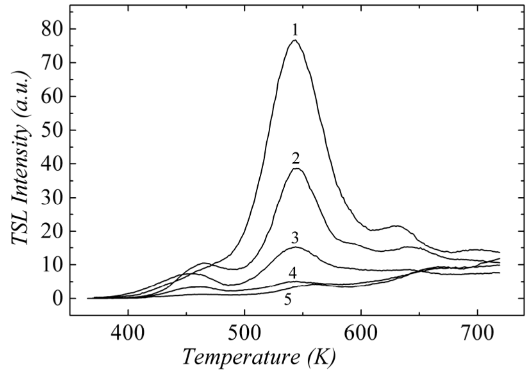

Figure 1. TSL curves of crystals irradiated in the WWR-SM reactor with neutron fluence: curve 1 - 2 × 10^{14} n/cm^2, 2 - 1 × 10^{16} n/cm^2, 3 - 5 × 10^{16} n/cm^2, 4 - 2 × 10^{17} n/cm^2 and 5 - 5 × 10^{17} n/cm^2.

To study the trends of the abovementioned drops, TSL intensities for peaks at 450 K and at 550 K were plotted as a function of the irradiation fluence (see curves 1 and 2 in **Figure 2(a)** respectively); the experimental points fitted well exponential curves.

The experimental results obtained after irradiation of the same crystal group in the reactor IBR are shown in **Figure 3**. The steady increase of peaks at 450 K (curve 2) and at 550 K (curve 1) is observed up to the dose of 10^{15} n/cm^2 and not until relatively high fluencies their complicated non-steady behavior is registered.

Thus, data presented in **Figures 2** and **3** are vivid illustration of the fact that in the same doped crystals (MgO:Mn^{2+}) different effects are provoked in the same neutron fluence ranges in the reactors differing by their parameters.

3.2. The Results of Step-by-Step Irradiation of Mgo:Mn^{2+} Crystals in the Both Reactors

The result of step-by-step irradiation of MgO:Mn^{2+} crystals in the both reactors is given in **Figure 4**. At first the crystal was irradiated in the IBR reactor, registration of TSL peaks following, *i.e.* short-term heating up to approximately 800 K with subsequent cooling, then the crystal was again irradiated this time in the WWR-SM reactor. Dependences 1 and 2 (**Figure 4**), corresponding to TSL peaks at 450 K and 550 K after final irradiation, show the reverse trend of the peaks behavior, whereas in the previous cases (**Figure 2** and **3**) the same trend is observed for the similar dependences. This result can be explained on the basis of the IR spectra trends (curves 3 and 4, **Figure 4**): the broad maximum of curve 1 should be caused by maximal accumulation of hole centers, responsible for the IR bands at 3290 and 1600 cm^{-1}, which is corroborated by the alternation of curves 3 and 4

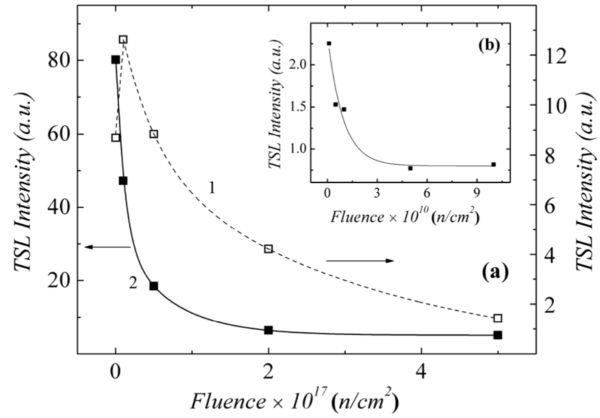

Figure 2. (a) Dependence of TSL intensity for peaks at 450 K (curve 1) and 550 K (curve 2) on neutron fluence in the WWR-SM; (b) Dependence of TSL intensity for peak at 550 K on neutron fluence (a generator) according to data of paper [3].

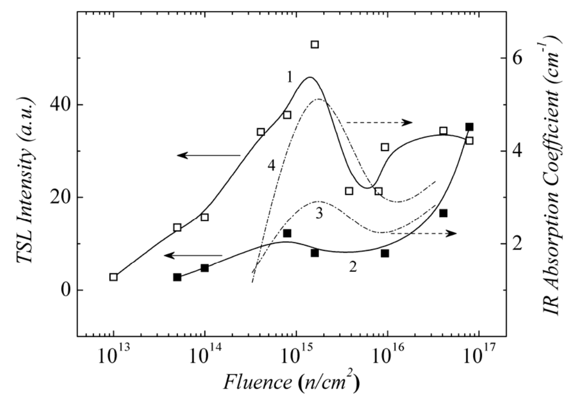

Figure 3. Dependences of TSL intensity for peaks at 550 K (curve 1) and 450 K (curve 2) as well as of IR absorption coefficient at 3290 and 3370 cm^{-1} (curve 3) and 1600 cm^{-1} (curve 4) on fast neutron fluence for crystals irradiated in the IBR-2 reactor.

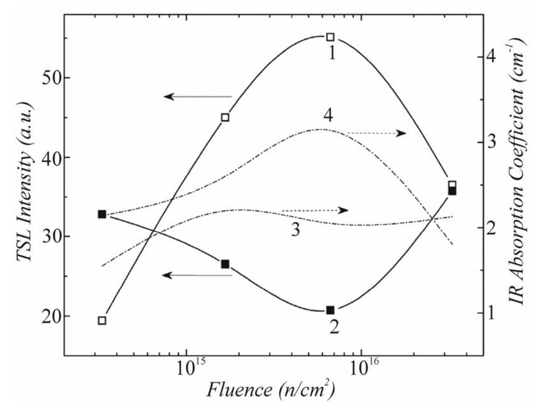

Figure 4. Dependences of TSL intensity for peaks at 450 K (curve 1) and 550 K (curve 2) as well as of IR absorption coefficient at 3290 and 3370 cm^{-1} (curve 3) and 1600 cm^{-1} (curve 4) on fast neutron fluence for crystals irradiated step-by-step in the IBR-2 and WWR-SM reactors.

maxima in the fluence range under study.

As to curve 2 minimum, it is due to the general decrease of hole centers amount after repeated irradiation of samples in the WWR-SM reactor (compare intensities of curves 3 and 4 in **Figure 4** with intensities of curves 3 and 4 in **Figure 3**), which caused deficiency of defects forming trapping centers of more complicated configuration that give rise to peak at 550 K.

4. Discussion

TSL intensity decrease in $MgO:Mn^{2+}$ (the corresponding dependence is shown in **Figure 2b**) is identified as a "small dose effect" [3]. It is known that this effect is not limited to a specific dose range and in different cases, differing doses can operate as small ones. The essence of the effect [4] is the qualitative change of radiation-induced modification of the material properties after some range of "small doses". Under irradiation apart from generation of primary radiation defects, rearrangement of existing defects can take place, up to their removal from the crystal (for instance, interstitial atoms emerge on the surface, are trapped by dislocations, annihilate with holes etc.). The result depends not only on the fluence but on the state of the crystal also.

Usually the concentration of initial defects (n) in the crystal is by several orders of magnitude less than the concentration of sites (N), so the efficiency of the initial defects rearrangement is considerably less than the efficiency of radiation defects generation. Nevertheless, in the case of the crystal heavily defected initially, when n/N is not very small, the efficiency of rearrangement and annihilation of the initial defects will be more than the efficiency of radiation defects generation, particularly under the action of particles with relatively small energies [4], that is perfection of the crystal occurs.

In our opinion the abovementioned situation can be successfully implemented in $MgO:Mn^{2+}$ crystal (having initial defects of growth and dope character [5]) in the WWR-SM reactor under the appropriate irradiation conditions, namely considerably low (as compared with thermal neutrons) flux of fast neutrons, that are necessary for creation of new structural defects in oxides, and high gamma background. Therefore, at the starting stage of irradiation (2×10^{14} n/cm^2) hole trapping centers become more apparent because the dominating intrinsic defects in the unprocessed original crystals of alkaline earth metals oxides are cation vacancies [6]. TSL peaks at 450 and 550 K on curve 1 (**Figure 1**) are due to these hole centers.

With neutron fluence growth, until the process of radiation defects generation becomes dominating, increasingly more of the irradiation energy absorbed by crystal is spent on the rearrangement of the existing defect structure and destruction of the trapping centers, which in its turn leads to TSL intensity decrease. In our case, this process lasts up to the fluence of 5×10^{17} n/cm^2 (curves 2 - 5, **Figure 1**).

Such trend of the processes in the irradiated $MgO:Mn^{2+}$ crystal *i.e.* destruction of hole-type trapping centers at the starting stage of the irradiation, is confirmed by dependence of the absorption coefficient peak of IR bands at 3290, 3370 and 1600 cm^{-1} on the neutron fluence (**Figure 5**).

The first two bands are caused by stretching vibrations of OH^- groups in the appropriate environment (O^{2-} - [Mg^{2+}] - OH^- and OH^- - [Mg^{2+}] - OH^-). The exact model of centers giving rise to the band at 1600 cm^{-1} is not ascertained at present. This band may be attributed to hydroxyl-containing centers as in many cases it behaves similarly to bands at 3290 and 3370 cm^{-1} [7].

It is possible to explain the results shown in **Figure 3** taking into account irradiation conditions in the IBR reactor. In this reactor fraction of fast neutrons is higher than in the WWR-SM reactor and gamma background is not very high, *i.e.* conditions for intensive displacement of ions in magnesium oxide are favourable. That is the cause why for crystals, irradiated by moderate neutron fluencies ($\leq 10^{15}$ n/cm^2), the concentration of detected complex hydroxyl-containing centers of V_{OH} type increases (curves 3 and 4, **Figure 3**) as well as TSL peak intensity does (curves 1, 2). After accumulation of definite amount of lattice imperfections (effect of doping Mn [5] plus atom displacements caused by fast neutrons) the situation, changing the trend of the processes in the irradiated crystal, is brought about (when n^*/N is not very small, where n^* is the total amount of defects). These processes are accompanied by the explicit characteristic of the "small dose effect"—nonsteady trend of curves

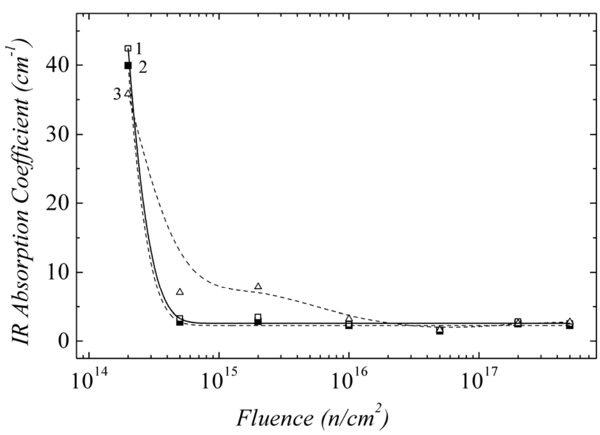

Figure 5. Dependences of IR absorption coefficient at 3290 cm^{-1} (curve 1), 3370 cm^{-1} (curve 2) and 1600 cm^{-1} (curve 3) on neutron fluence for crystals irradiated in the WWR-SM reactor.

with further fluence increase [3].

5. Conclusions

- The "small dose effect" has been detected in TSL glow curves and IR spectra of doped $MgO:Mn^{2+}$ crystals for moderate neutron fluencies (10^{14} - 10^{17} n/cm^2).
- Under the invariable conditions of irradiation (the intensity and spectrum) TSL peaks at 450 and 550 K for $MgO:Mn^{2+}$ crystals are changing with the fluence increase in the same mode, which indicates the similar nature of corresponding trapping centers.
- Correlation between the high-temperature TSL curves and IR absorption spectra for $MgO:Mn^{2+}$ doped crystals irradiated in the different reactors shows that nature and behavior of TSL peaks at 450 and 550 K are mainly determined by hydroxyl containing hole centers.

6. Acknowledgements

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7. References

[1] S. Dolgov, T. Kärner, A. Lushchik, A. Maaroos, N. Mironova-Ulmane and S. Nakonechny, "Thermoluminescence Centers Created Selectively in MgO Crystals by Fast Neutrons," *Radiation Protection Dosimetry*, Vol. 100, No. 1-4, 2002, pp. 127-130.

[2] V. Kvachadze, G. Dekanozishvili, T. Kalabegishvili, V. Vylet, M. Galustashvili, Z. Akhvlediani, N. Keratishvili and D. Zardiashvili, "Thermally Stimulated Luminescence of MgO Ceramic and Single Crystal Samples Irradiated in Reactor," *Radiation Effects and Defects in Solids*, Vol. 162, No. 1, 2007, pp. 17-24. doi:10.1080/10420150601016235

[3] V. Kvatchadze, G. Dekanozishvili, M. Abramishvili, Z. Akhvlediani, M. Galustashvili and T. Kalabegishvili, "Thermoluminescence and Absorption Spectra of MgO Crystals Irradiated by Fast Neutron Fluxes of Low Intensity," *Physica Status Solidi* (*A*), Vol. 206, No. 11, 2009, pp. 2581-2585. doi:10.1002/pssa.200925016

[4] R. Atabekyan, V. Vinetskii, V. Gevorkyan, R. Ezoyan and G. Eritsyan, "Optical Small Dose Effect in the Radiation State Physics," *Pis'ma v Zhurnal Tekhnicheskoi Fiziki*, Vol. 9, No. 23, 1983, pp. 1448-1451.

[5] M. G. Abramishvili, K. A. Garibashvili, A. I. Kurasbediani, V. G. Kvachadze, Z. G. Akhvlediani, M. G. Galustashvili, T. L. Kalabegishvili and S. Sobolevskaya, "The Optical and Paramagnetic Absorption Spectra of MgO:Mn Single Crystals Irradiated in a Reactor," *Radiation Effects and Defects in Solids*, Vol. 159, No.10, 2004, pp. 543-550. doi:10.1080/10420150412331326921

[6] P. Kofstad, "Deviation from the Stoichiometry, Diffusion and Electrical Conduction in the Simple Metallic Oxides," Mir, Moscow, 1975.

[7] M. G. Abramishvili, Z. G. Akhvlediani, M. G. Galustashvili, T. L. Kalabegishvili, A. I. Kurasbediani and V. G. Kvachadze, "Radiation-Induced Alterations of MgO: IR Absorption Spectra in a Wide Interval of Wave Numbers," *Radiation Effects and Defects in Solids*, Vol. 160, No. 6, 2005, pp. 237-224. doi:10.1080/10420150500275338

