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Thermoluminescent processes of $MgAl_2O_4$ irradiated at room temperature

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Two non-first-order kinetic peaks at about 350 and 500 K appear in the thermoluminescence (TL) spectra of single-crystal and polycrystalline $MgAl_2O_4$ irradiated with ionizing radiation at room temperature. Among others, emission from Cr^{3+} and Mn^{2+} ions are observed. TL measurements at different wavelengths indicate that the emitting centers have different recombination probabilities. Thermally-stimulated-current and thermoelectric-power measurements, together with results on the TL excitation spectra with ultraviolet radiation, indicate that both processes are due to thermally released electrons from traps. The V centers also act as recombination centers leading to a complex emission band at around 4.9 eV. Experimental results and numerical simulations based on a recently proposed mathematical model suggest that the massive presence of lattice defects, as a result of the antisite disorder and the nonstoichiometry of synthetic spinel crystals, may have a strong influence on electron-trapping and -detrapping processes, leading to complex-kinetics TL phenomena.

I. INTRODUCTION

It is now quite well established that ionizing irradiation induces charge rearrangement processes involving impurities and lattice defects in oxides such as MgO and Al_2O_3. Hence, thermoluminescence (TL) and thermally-stimulated-current (TSC) processes observed when heating these irradiated materials arise from charge carrier release from traps. So these studies provide information about crystal lattice defects and their interactions. In particular, the study of ionizing irradiation effects in $MgAl_2O_4$ spinel may lead to a better understanding of the high resistance of this material to displacing radiation damage[1,2] which makes it a good candidate for some fusion technology applications.

In the perfect $MgAl_2O_4$ spinel cubic structure, divalent cations occupy tetrahedral sites while trivalent cations are in octahedral sites. However, extensive cation antisite disorder (up to 25% site mixing) occurs in synthetic crystals, as has been detected by various techniques.[3–7] This results in local positive and negative charges which may act as electron- and hole-trapping centers. On the other hand, $MgAl_2O_4$ crystals are usually slightly nonstoichiometric with a certain percentage of excess Al_2O_3 molecules, cation vacancies being formed for charge compensation of the extra Al^{3+} ions. Some calculations and measurements seem to indicate that these vacancies are in octahedral sites[8,9] because the additional Al^{3+} ions occupy magnesium positions. Nevertheless, some recent calculations show that both types of vacancies may appear due to the nonstoichiometry.[10] This would be reflected on the kinetics of charge trapping and detrapping processes which are responsible for TL and TSC.

Only a few papers on TL of $MgAl_2O_4$ have been published to date.[11–15] Several peaks appear in the TL spectrum between 300 and 700 K, but results differ among the different works. A commonly observed glow peak at around 350 K has been ascribed to electron release from traps, which might be $[Al]^+_{Mg}$ centers (an Al ion at a Mg site).[16] Recombinations of these electrons at V and $[Mg]^0_{Al}$ (a Mg ion at an Al site, with a trapped hole) centers are proposed to be responsible for light emission of 4.8 and 4 eV, respectively.[15] One or two more TL processes occur at higher temperatures which have been attributed to either electron[13,14] or hole[11,12,15] release, light emission being due to several impurities, such as Cr, V, Fe, and Mn. The peak temperatures are different for each emission band,[13–15] perhaps due to a different charge capture cross section for each recombination center.[15] TL peaks also seem to be composite because their maxima shift to higher temperatures when a partial preheating run is made prior to the TL measurement.[15] Due to the rather complex shape of the glow curves, the existence of a quasicontinuous spectrum of electron and hole traps has also been proposed.[14] Some TSC measurements show that the first TSC peak appears at the same temperature as the first glow peak, but the other current peaks do not coincide with any glow peak.[12,15]

As seen above, the published results seem to show a somehow puzzling picture about TL processes in $MgAl_2O_4$ above room temperature. A possible influence of the different sample origins is not clear. One would also expect that the massive presence of defects related to the antisite disorder and the nonstoichiometry of samples would have some specific effect on TL process kinetics. For these reasons, a detailed study of TL in this material above room temperature has been made. Single-crystal and polycrystalline samples of different origins have been used. TSC and thermoelectric-power measurements have been made in order to determine which type of electric carrier is released in each process.

In a previous work,[17] two superimposed electron paramagnetic resonance (EPR) bands at $g=2.011$ and a broad composed optical absorption band at 3.4 eV have been related to V-type centers in $MgAl_2O_4$ x-ray irradiated at 90 K. These centers annihilate in a single step between 220 and 575 K. It has been proposed that this very wide annealing temperature range is due to the presence of a high-concentration electron trap. Therefore, optical absorption and EPR spectra have also been obtained in $MgAl_2O_4$ irradiated at 290 K to ascertain whether these types of centers are related to TL processes above room temperature.

II. EXPERIMENT

1-mm-thick samples from three different origins have been employed. Single-crystal samples (labeled UC samples) grown by the Czrochalski method have been purchased from Union Carbide. Some of them from a different batch were kindly supplied by Dr. R. Heidinger (Kernforschungszentrum Karlsruhe, GFR), as well as the polycrystalline transparent samples from Raytheon (USA) (labeled *R* samples) also used in this work, which have been fabricated by the fusion casting method.[18] Finally, highly Mn-doped Verneuil grown single crystals (labeled TG samples) were also obtained from Talla Gem (Spain). All samples were characterized by x-ray fluorescence to know their stoichiometry. Chemical analyses were also made to obtain their impurity content. Results are shown in Table I. Cr (in UC and *R* samples) has clearly been detected by photoluminescence measurements.

X-ray irradiations were performed at temperatures varying between 290 and 315 K through a 2-mm-thick aluminum plate at about 3×10^3 R min^{-1} with a Siemens tube (tungsten anode) operated at 50 kV and 30 mA. A ^{60}Co source was used for γ irradiations at a dose rate of 3.9 Mrad h^{-1}, and a 450-W Xe lamp was used for ultraviolet (uv) irradiations at different wavelengths.

TL measurements between 300 and 700 K have been made at 0.17 K s^{-1} with an apparatus described elsewhere.[19] EMI 9659 B and EMI 6256 S photomultipliers have been used. A motor-driven Bausch and Lomb monochromator inserted between the sample and the photomultiplier has been employed for recording emission spectra continuously by performing repetitive scans (one each 20 s) during a heating run. The measured data are sent to a Hewlett-Packard computer-controlled data acquisition system. The emission spectra have been corrected by the photomultiplier standard response. So this system allows us to obtain, from these corrected data taken from a single heating run, emission spectra at different temperatures; monochromatic glow curves at any wavelength and, by integrating data, the total light TL spectrum. In this way the "arbitrary units" here used for light intensity are always consistent among the different spectra and proportional to the emitted photon intensity, even in the case of the total light TL spectra, which is not the case when the photomultiplier current is plotted directly against temperature.

TSC measurements were made in a chamber mounted in the beam line of a HVEC Van de Graaff accelerator.[20] About 1000 V cm^{-1} were applied to the platinum electrodes evaporated on the sample. Optical absorption spectra were obtained with a Cary 17 spectrophotometer. EPR measurements were made at the Universidad Autónoma de Madrid with a Varian E-12 spectrometer working in the *X* band. Thermal annealing of the EPR signals was performed by heating the sample up to a given temperature for 3 min in a Varian E 257 variable temperature accessory and measuring the spectra at 300 K.

III. RESULTS

A. TL and TSC measurements

TL spectra of $MgAl_2O_4$ γ irradiated at 290 K are shown in Fig. 1. Two peaks are clearly seen for the three different sample types. UC samples have their maxima at around 355(I) and 495(II) K, *R* samples at 355 and 515 K, and TG samples at 335 and 515 K. The relative intensities are different but these spectra seem to indicate that the same two glow peaks are observed in each type of sample. Both peaks are very wide, the high-temperature half-width of the first peak is larger than that of its low-temperature counterpart, while the second peak is almost symmetric.

X-ray and uv irradiations causes the same TL spectrum as γ irradiation in all samples, although with different relative peak heights. This is due to the different doses and dose rates of the three irradiation sources. Peak I shifts to higher temperature when the irradiation temperature is increased. The dependence of the TL response on the photon energy for uv irradiation is shown for *R* samples in Fig. 2, in which the peak-II height divided by the number of incident photons has been plotted against the photon energy. The same spectrum has been obtained for peak I. As seen in this figure, a band at 5.3 eV and the tail of another one at energies higher than 6.2 eV are clearly observed. This agrees very well with the optical absorption spectrum of as received *R* samples. In UC samples, where no optical absorption band at 5.3 eV appears, the TL excitation spectrum only shows the high-energy tail.

Figure 3 shows the emission spectra for *R* samples. Bands at 1.77 (700 nm), 2.4 (515 nm), and 3.0 (410 nm) eV are common for both glow peaks. The 1.77-eV band is a complex one, as shown in the inset of this figure. A very weak emission at around 2.0 eV (620 nm) also appears at high temperature. This 1.77-eV emission has the same

TABLE I. Stoichiometry [x in $MgO\cdot x(Al_2O_3)$] and main impurity content (ppm) of $MgAl_2O_4$ samples.

Sample type	x	Fe	Ti	Cr	Mn	V
UC	1.02	100	50	<20	10	10
R	1.1	35	30	<20		
TG		<15		<20	15000	

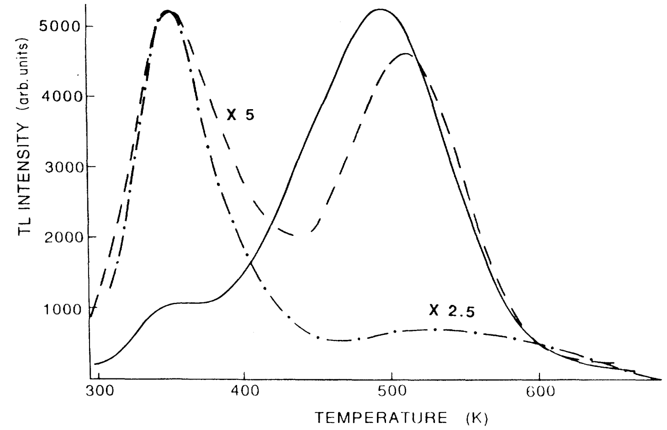

FIG. 1. TL spectra of MgAl$_2$O$_4$ γ-ray irradiated up to 650 krad: UC (——), *R* (— — —), and TG (—·—·—·) samples.

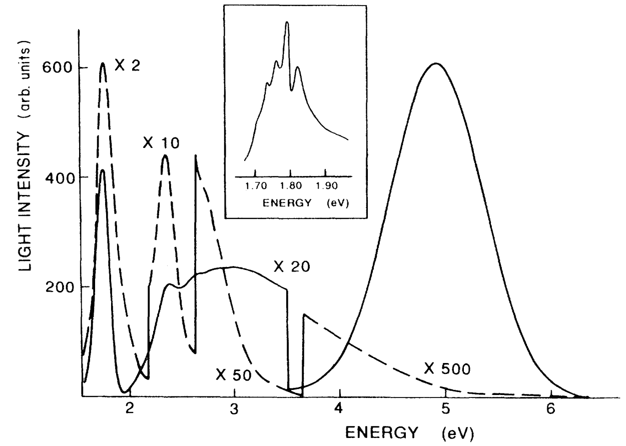

FIG. 3. Emission spectra for peaks I (——) and II (— — —) in *R* samples. The inset shows the emission spectrum around 1.80 eV measured with a higher wavelength resolution.

structure as the well-known Cr^{3+} emission.[21] This emission has also been detected by photoluminescence measurements with a JY3CS Jobin-Yvon spectrofluorimeter. Excitation and emission spectra agree very well with those reported for octahedral Cr^{3+} ions in the literature.[21-24]

In addition to these bands, an emission peaked at about 4.9 eV (250 nm) is dominant in the first glow peak but does not appear in the second one. Its maximum shifts to lower energies with increasing temperature and a shoulder can be observed, showing that it is actually composed by two strongly overlapped bands (Fig. 4). These two bands peaked at 4.45 (280 nm) and 5.1 (245 nm) eV have also been observed in radioluminescence (RL) measurements under electron irradiation in our laboratory. Their relative intensities vary with temperature between 150 and 600 K. Since they are clearly observed in RL measurements in the temperature range in which the second TL peak appears, the fact that no uv emission band is observed in this TL process may be explained by assuming that the corresponding recombination centers have disappeared before.

The same emission bands as in *R* samples appear in UC samples. The 1.77-eV band is more intense and the 2.05-eV one is much weaker. Also, the ultraviolet emission peaks at higher energy (about 5.2 eV); this could be due to the relative intensities of both uv bands being different than in *R* samples.

TG samples only show two emission bands, a weak one around 5.0 eV (238 nm) and a high-intensity one with maximum at 2.4 eV (515 nm). Photoluminescence studies of this last one in as-received TG samples show that this emission and its excitation spectrum are the same as those reported for tetrahedral Mn^{2+} luminescence.[25,26]

TL spectra at those wavelengths corresponding to the maximum of each emission band (monochromatic TL recordings) are plotted in Figs. 5 and 6 for UC and *R* samples, respectively. They confirm the above-mentioned correspondence among glow peaks and emission bands. However, the second TL peak maximum appears at a different temperature value for each emission band, as it was previously observed,[13-15] and it is sample-type dependent. Its shape and half-width, and thus the form

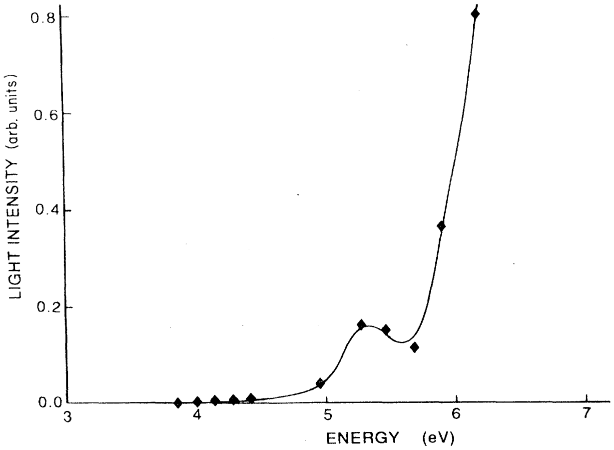

FIG. 2. Ultraviolet excitation spectrum of the TL of *R* samples.

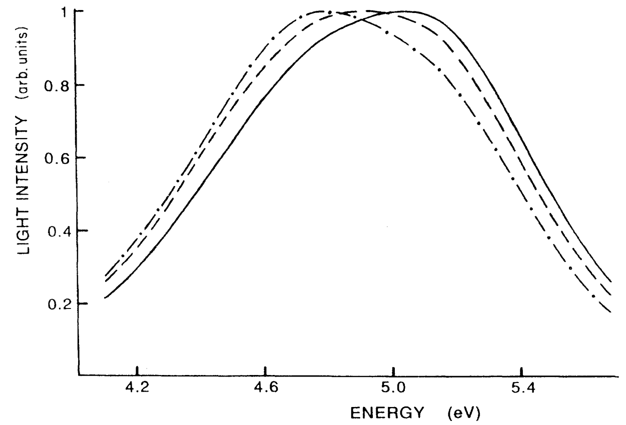

FIG. 4. Normalized ultraviolet emission band at 300 (——), 325 (— — —), and 350 (—·—·—·) K in *R* samples.

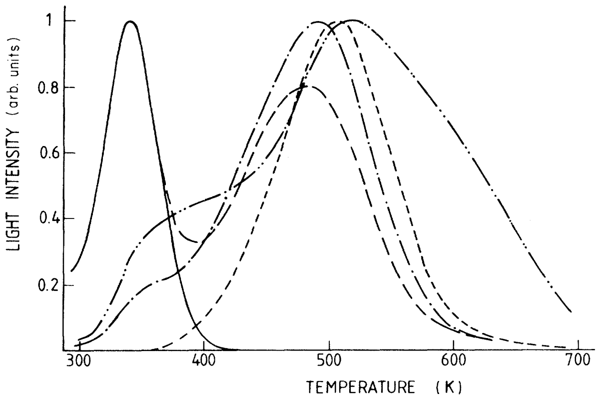

FIG. 5. Normalized TL spectra of UC samples measured at different wavelengths: 4.95 (——), 3.0 (— —), 2.4 (—··—), 2.0 (— — —), and 1.77 (—·—) eV.

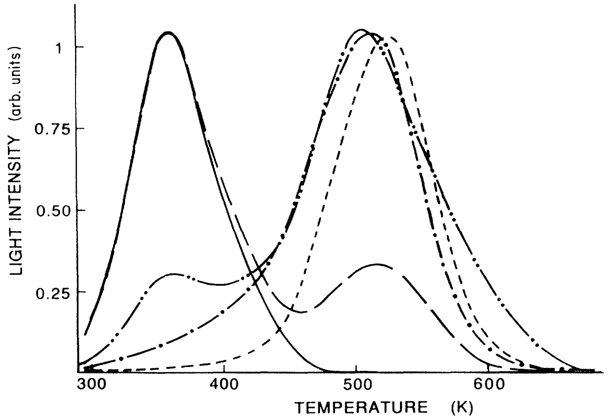

FIG. 6. As in Fig. 5 for *R* samples.

factor (i.e., the ratio of the high-temperature half-width to the total half-width), also depend on the emitted light wavelength, although for each emission the form factor is the same for both the UC and *R* samples. These data are summarized in Table II for a saturating dose value (see below).

The dependence of the TL spectrum on the irradiation dose is somehow complicated.[27] Peak II becomes narrower and shifts to lower temperatures with increasing dose, at it can be seen for *R* samples in Fig. 7, where the TL curves have been normalized to the peak-II height for the sake of clarity. The light intensity for all emissions in this peak shows saturating growth. The dose dependence of peak position and half-width is different for each emission wavelength, as can be seen in Fig. 8.

The peak position and the half-width for each emission in peak I seem not to depend on dose. Light intensities grow with saturating trends for all emissions but the 4.9-eV one, whose increase is slightly supralinear at least up to 1000 krad. UC samples have not been studied in such a detail as *R* samples, but the available results indicate that both types of spinel samples behave in a similar way.

Isothermal light decay (phosphorescence) measurements at different wavelengths have been obtained at several temperatures. They show a nonusual behavior. For a given temperature, the decay rate of each emission band is different from the others. They do not follow any clear kinetics order. Also, for a given wavelength the decay rates cannot be ordered as it would be expected, i.e., showing an increase in the decay rate when the temperature is increased, as can be seen in Fig. 9. A typical first-order or second-order kinetics analysis leads to curve decomposition into three or more components, which are different for each wavelength and which do not show any clear ordering with temperature. This seems not to have any physical meaning. Other attempts to fit the phosphorescence curves by different methods[28] failed in giving any clear result. The most interesting results were obtained with the method by May and Partridge,[29] which directly calculates the kinetics order parameter: it was found that this decreases continuously along the phosphorescence measurement time for each curve. Moreover, for a given wavelength, the obtained values decrease when the temperature increases, being, however, different for the distinct emission wavelengths.

The TSC spectra of UC and *R* samples electron irradiated at 290 K are plotted in Fig. 10, in which the curves represent the difference between the spectra obtained after and before irradiation in both types of samples. A current peak at about 320 K, whose asymmetric shape is similar to that of glow peak I, and another one at around 630 K are clearly seen. The difference of about 35 K between the first TSC peak maximum and that of the first TL peak can be explained as due to the lower heating rate used in these measurements (because of experimental reasons), which was 0.07 K s.$^{-1}$ So, it is sensible to assume that both the 355-K glow peak and the 320-K current peak are caused by the same process. Thermoelectric power measurements, made by maintaining a temperature gradient across the sample during the heat-

TABLE II. Height, position, half-width, and form factors of glow peak II for each emission band in the UC and *R* samples, γ-ray irradiated up to 650 krad.

	Height (a.u.)		Position (K)		Half-width (K)		Form factor	
Emission (eV)	UC	*R*	UC	*R*	UC	*R*	UC	*R*
3.0	12	15	480	515	125	100	0.48	0.48
2.40	60	20	520	500	185	120	0.59	0.59
2.0	220	82	510	525	105	90	0.44	0.45
1.77	1200	370	490	510	120	100	0.42	0.43

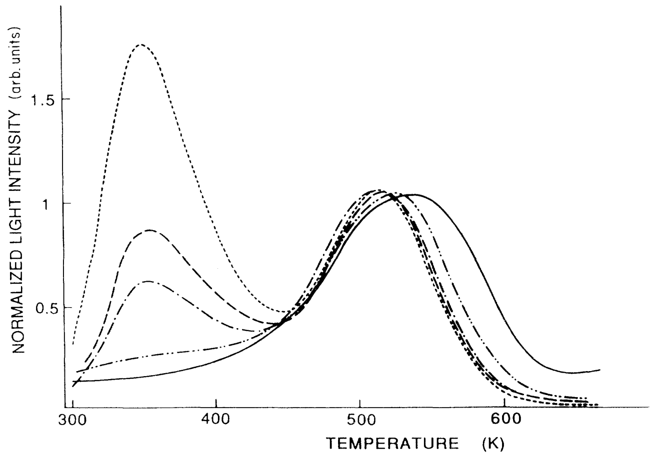

FIG. 7. TL spectra of $MgAl_2O_4$ (*R* samples) x-ray irradiated up to 3 (——), 6 (—··—), 60 (—·—·—·), 180 (— — —), and 860 (· · · ·) krad. Light intensities have been normalized to peak-II height.

ing run, show that the cold sample side develops a negative voltage with respect to the hot side in the whole temperature range between 300 and 700 K, thus indicating that electrons are thermally released in the range where both glow peaks appear. This supports that there is also charge release in the second TL process.

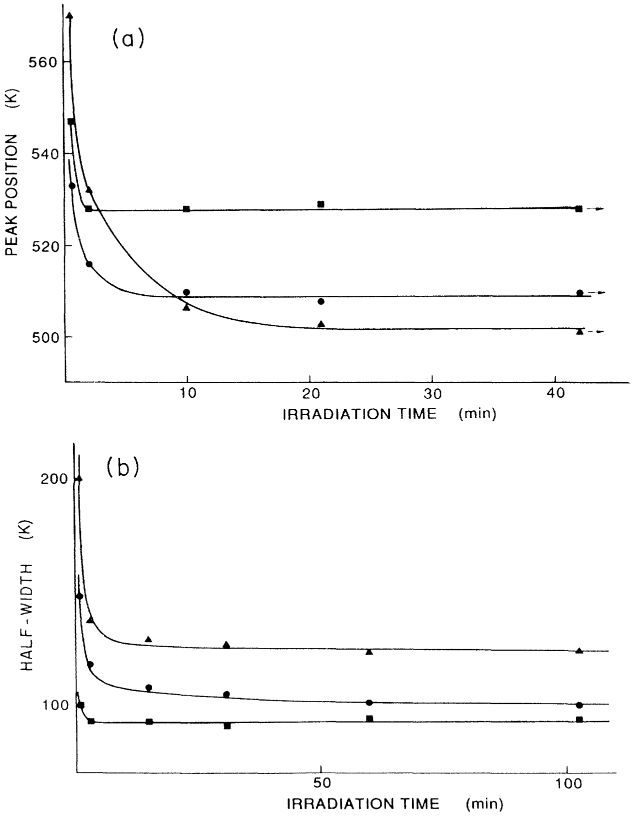

FIG. 8. Dependence on the x-ray irradiation dose of peak-II position (a) and half-width (b) for the emissions at 2.4 (▲), 2.0 (■), and 1.77 (●) eV in *R* samples.

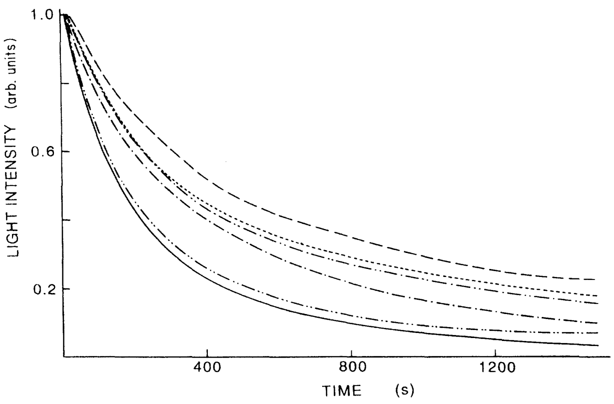

FIG. 9. Normalized isothermal light decay curves at 1.77 eV at different temperatures: 446 (· · · ·), 466 (—··—·—), 481 (—··—··—), 493 (—·—·—·), 511 (— — —), and 530 (——) K.

B. Optical absorption and EPR measurements

As-received UC samples show the EPR spectrum of Cr^{3+} ions in octahedral position.[30,31] The hyperfine structure of the EPR spectrum of Mn^{2+} ions[32,33] is also observed in UC and TG samples. In the last ones, five narrow optical absorption bands at 2.6, 2.79, 2.92, 3.25, and 3.4 eV, which have been ascribed to Mn^{2+} ions in tetrahedral sites,[25,26] are also clearly seen. In the single-crystalline UC samples, two strong optical absorption bands at 4.8 and 6.4 eV, as well as a very weak one at 0.62 eV, appear. The two first ones agree in wavelength with charge-transfer bands related to octahedral Fe^{3+} ions,[11,34,35] but they have not been detected by EPR. The 0.62-eV band is due to tetrahedral Fe^{2+}.[22] Finally, as-received *R* and TG samples show an absorption band at about 5.3 eV which might be related to *F* centers,[36,37] which can be formed during crystal production depending on the growing conditions.[38,39]

On irradiation, the Cr^{3+} EPR signal vanishes, while the Mn^{2+} concentration remains constant. The Cr^{3+} recovery occurs on heating the sample between 300 and

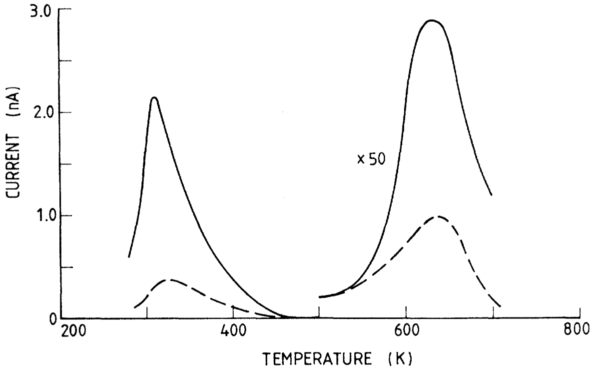

FIG. 10. TSC spectra of UC (——) and *R* (— — —) type $MgAl_2O_4$ samples irradiated at 290 K.

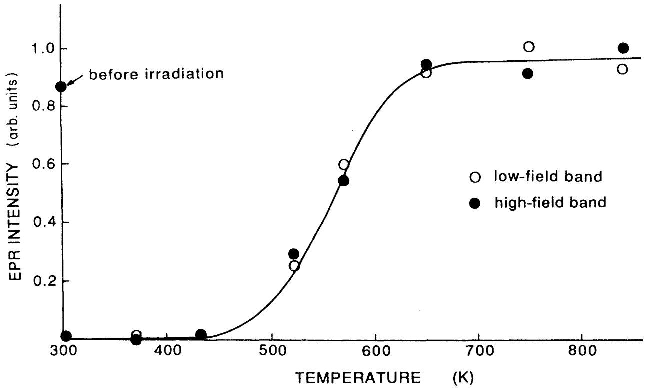

FIG. 11. Thermal recovery of the Cr^{3+} EPR band after x-ray irradiation at 290 K in UC samples. The intensity has been normalized to the maximum value.

700 K, as can be seen in Fig. 11.

X-ray irradiation at room temperature also induces in UC and *R* $MgAl_2O_4$ samples a broad composed optical absorption band between 2.5 and 5 eV peaked at about 3.3 eV, and two superimposed EPR spectra, labeled EPR1 and EPR2, centered at $g = 2.011$. They have been ascribed in a previous work to different *V*-type centers.[17] Our results indicate that a high *V*-center concentration is generated by ionizing irradiation, as a consequence of the nonstoichiometry of spinel crystals. In addition, an optical band at about 5.8 eV is also induced by x-ray irradiation at 290 K.

A pulsed thermal annealing method has been used to study the thermal stability of *V* centers. After a heating pulse up to a given temperature value, the optical absorption and the EPR spectra are measured at room temperature. It has been found that all *V* centers decay between 350 and 570 K in a broad single step (Figs. 12 and 13). All optical absorption bands between 2.5 and 5 eV behave as the 3.3-eV one plotted in Fig. 12. During thermal annealing of irradiated samples, a new EPR narrow line at $g = 2.01$ (width less than 20 G), labeled EPR-3, appears. It grows until 550 K and then decays between 550 and 800 K (Fig. 13). Its thermal evolution is similar to that of the 5.8-eV optical absorption band (Fig. 12), at least between 450 and 700 K.

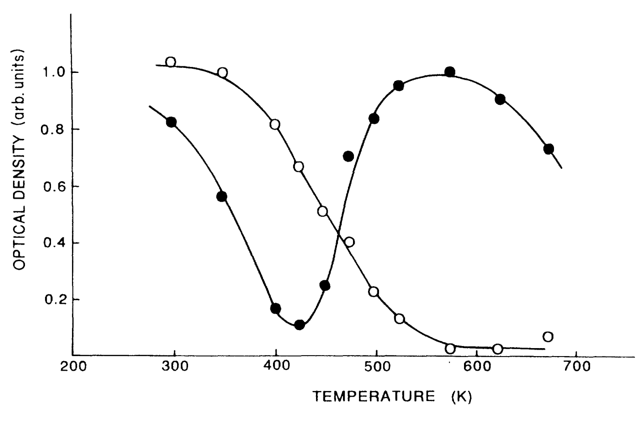

FIG. 12. Thermal annealing curves of the 3.3- (○) and 5.8- (●) eV optical absorption bands in x-ray-irradiated UC-type $MgAl_2O_4$ samples. The optical densities have been normalized to their maximum values.

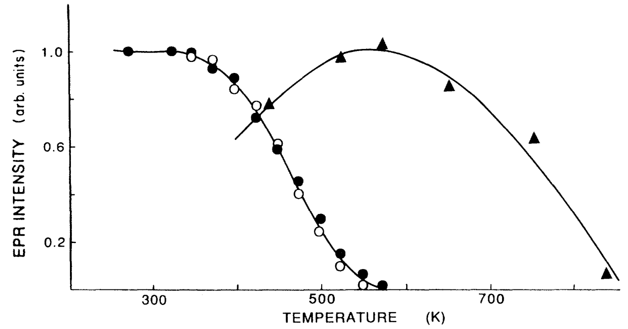

FIG. 13. Thermal annealing curves of the EPR-1 (○), EPR-2 (●), and EPR-3 (▲) bands in x-ray-irradiated UC-type $MgAl_2O_4$ samples. The EPR intensities have been normalized to their maximum values.

IV. DISCUSSION AND CONCLUSIONS

It has been shown that there is a TSC peak which coincides with the first TL peak, so it is straightforward to conclude that this is due to thermal release of charge carriers from traps. Thermoelectric power measurements indicate that there is charge release not only in the temperature range where the first glow peak appears, but also in that of the second peak. Moreover, these measurements indicate that the charge carriers must be electrons. That both TL processes arise from thermal release of the same type of electric carriers is also supported by other experimental results. Both peaks have several common emission bands, so they have common recombination centers. Also, they are simultaneously generated by uv light having the same excitation spectrum. The fact that this spectrum shows a band at 5.3 eV, which has been related to *F* centers[36,37] in as-received samples whose optical absorption spectrum has a band at this energy, suggests that illumination in this band induces electron capture at traps responsible for both glow peaks.

The lack of a TSC peak at the temperature of the second TL peak has also been observed by other authors[12,15] who obtained another TSC peak at higher temperatures, as has been observed here. They ascribed the first TL peak and both TSC peaks to electron release and the second TL one to hole release. The nonobservation of a TSC peak coinciding with the second TL process was explained in terms of the difference between electron and hole mobilities and between hole capture cross sections by different impurities.[12] In another study,[15] the high-temperature TSC peak seemed to be composed and might have a contribution due to electron release which makes only a small contribution to TL. Our results are not consistent with these models since they indicate that both TL processes are due to electron release from traps. An alternative explanation for the difference in position between the high-temperature TL and TSC peaks will be given below.

The emission band at 1.77 eV is due to octahedral Cr^{3+} emission by electron capture at Cr^{4+}, as was stated above. This agrees with earlier results[11,12,14,15] and is also consistent with the irradiation-induced vanishing of the Cr^{3+} EPR signal and its recovery by heating. The 2.4-eV band may be ascribed to Mn^{2+} emission, as indicated by the photoluminescence measurements in the Mn-doped TG samples. Some authors have related it to emission from V^{3+} or Fe^{2+} ions.[11,12,15] This emission must be induced by energy transfer from a neighboring defect because the heights of the Mn^{2+} optical absorption, EPR, and photoluminescence spectra do not change either by irradiation or during the heating run. The nature of the recombination centers responsible for the 3.0- and 2.0-eV bands cannot be known from our results.

The 4.9-eV emission, which is actually composed of two bands at around 4.5 and 5.1 eV, is likely not related to impurities because of its nonsaturating behavior with dose. These two bands could arise from electron-hole recombination at different types of *V* centers, since they only appear while *V* centers are still present in the crystals. This has been suggested for an emission at 4.8 eV by White *et al.*[12] (although this has also been ascribed to recombination with holes trapped at $[Mg]_{Al}^{-}$ centers).[15] The existence of at least two different types of *V* centers, as revealed by EPR measurements,[17] supports the conclusion that each of these uv emissions band is associated with a different *V* center. A radioluminescence emission at 4.95 eV in MgO has been attributed to the same type of recombination.[40] The nonsaturating behavior of these emissions, at least up to the dose values reached here, must be a consequence of the existence of a large amount of trapping and recombination centers. Our results indicate that this is true for the last ones, as it is indicated by the *V*-center EPR spectrum intensity.

The differences among monochromatic TL curves concerning the temperatures at the glow peak maxima and the peak half-widths can be explained as due to differences in the electron capture cross section for each recombination center, as has been previously suggested.[15] This also would explain the different decay rates found for the isothermal monochromatic light decay curves at a given temperature value.

The shift of the second glow peak to lower temperature with increasing dose, i.e., with increasing trap filling, is typical of a non-first-order kinetics process. Its shape also indicates that its kinetics order is far from 1. This seems to also be true for peak I because of its asymmetric shape and of the supralinear dependence on dose of its main emission, according to the mathematical predictions by Kantorovich *et al.*[41,42] This conclusion is supported by the observed shift of peak I to higher temperature for increasing irradiation temperature: since this is close to the peak maximum, trap filling for this peak is simultaneous to trap emptying during irradiation, hence, the higher the irradiation temperature the larger the emptying rate, this resulting in a lower concentration of trapped electrons for a given dose. These non-first-order processes are responsible for the preheating effects observed by Lorincz *et al.*[15] who attributed them to glow peaks being composed. These effects would actually be due to partial emptying of traps by the heating pulse prior to the TL measurement.

Qualitative numerical simulations of the TL complex behavior have been made by solving the differential equation system proposed by Bull *et al.*[43] using a variable order and step-width Runge-Kutta method, which was previously checked to give very good results for single first- and second-order kinetics peaks. Different arbitrary values for each recombination coefficient, as indicated by our experimental results, have been used. The presence of a shallow electron trap in a very high concentration has also been included. This assumption is based on the existence of antisite disorder in synthetic spinel crystals, which may provide such a high concentration of electron as well as hole traps. The $[Al]_{Mg}^{+}$ defect (an Al^{3+} ion occupying a Mg site) would be an effective electron trap, leading to the $[Al]_{Mg}^{0}$ center by electron capture during irradiation. The $[Mg]_{Al}^{-}$ defect (a Mg^{2+} ion at an Al site) would act as a hole trap. The nonstoichiometry of crystals is also responsible for the large cation vacancy concentration, as has been already discussed. This shallow electron trap does not need to be related to any specific TL process but, because of its high concentration, its electron-capture probability is high enough to induce the charge carriers released from other traps to be retrapped and released by it many times before the final recombination. This leads to processes whose kinetics order is very different from 1, which may well be changing along with the trap emptying process as phosphorescence measurements seem to indicate, thus this parameter has no clear physical meaning.

With the two above-mentioned conditions (i.e., the presence of a high-concentration shallow electron trap and the recombination probabilities being different for each recombination center), this mathematical model can qualitatively simulate all experimental features found in this work. The presence of a high-concentration trap explains the non-first-order kinetics behavior, as reflected by the glow peak parameter and their dependence on dose as well as by the phosphorescence results. The different recombination coefficients are also responsible for the time and temperature dependence of the kinetics parameter in the isothermal light decay measurements. They also lead to the differences found in monochromatic TL curves.

The mathematical model has also been used for qualitative simulations of trapping and recombination center thermal stability. Besides decreasing steps for the concentration of filled traps related to each TL process, an initial increase followed by a further annealing step for the high-concentration electron trap can be obtained for each charge release process. This resembles, at least partially, the behavior of the optical absorption band at 5.8 eV and the EPR-3 signal, suggesting that both might be related to $[Al]_{Mg}^{0}$ centers. More work is needed to ascertain this point. Another consequence of the presence of a high-density electron trap is a very wide annealing step for each recombination center, as it has been found for *V* centers in this work.

Finally, TSC curves have also been numerically simulated for different sets of parameters leading to different kinetics order values. It is shown that a TSC peak becomes wider and with lower intensity, as well as its maximum shifts to higher temperature, for simulated increasing kinetics order values. A similar effect is obtained when increasing the shallow electron trap concentration. Hence, the TSC peak related to the second TL peak can be very flat and far from this one, in agreement with our experimental results. This supports that the second TSC peak is related to the second TL one, and so both glow peaks have corresponding current peaks.

In summary, γ-ray-, x-ray-, or uv-irradiated $MgAl_2O_4$ samples of different origins show two TL processes at about 350 and 500 K, both due to electron release from traps. Besides the role of some impurities as emitting centers having different recombination probabilities for the released electrons, these TL processes are strongly influenced by the massive presence of lattice defects as a result from the antisite disorder and the nonstoichiometry of samples. This also leads to a high concentration of *V*-type centers acting as recombination centers.

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