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Luminescence from electron-irradiated sapphire

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Point defects have been produced by electron irradiation in single-crystal α-Al_2O_3 from two different sources. Time-resolved luminescence spectroscopy has been used to study emission spectra and atomic-displacement thresholds for the defects. An emission band at 3.8 eV is present in only one of the crystals while a band at 4.2 eV is observed in both crystals. The former is probably due to F^+-center emission and the latter possibly arises from an F-center transition. Emission kinetics are consistent with detrapping of electrons from two shallow traps.

I. INTRODUCTION

Interest in the many technological applications of sapphire (α-Al_2O_3) has inspired an extensive study of its properties in recent years. Sapphire is used as a lasing material in solid-state lasers (e.g., ruby, Cr-doped sapphire), and as a substrate for silicon microelectronic devices. Its thermoluminescence properties suggest use as an efficient radiation dosimeter.[1,2] The physical properties of sapphire, such as its high melting point (2310 K), hardness, extreme strength, and resistance to radiation damage, have led to serious consideration of this material as an insulator for the first wall of a nuclear fusion reactor.[3–6]

Studies of defects in sapphire, their production mechanisms, and emission characteristics have led to a greater understanding of the electrooptic properties of this material. Levy and Dienes[7] observed that an absorption band produced at 6.1 eV (204 nm) in sapphire by fast neutrons was not present under X or γ irradiation. They therefore suggested that this band must be associated with a lattice defect created by atomic displacement. Arnold and Compton[8] produced the same band at 77 K by fast-electron irradiation. They monitored the change in optical density as a function of incident electron energy, and were able to determine a threshold energy for atomic displacement by comparing their experimental data with the calculated cross sections for the two possible displaced ions, oxygen and aluminum. Best fits were obtained for a threshold value of 0.43 MeV, corresponding to a displacement energy (that is, the maximum energy transferable to a displaced ion) of 50 eV for aluminum or 90 eV for oxygen. In a further study,[9] they presented very similar data, and obtained a threshold of 0.37 MeV, giving displacement energies of 40 or 70 eV for aluminum or oxygen, respectively. While the authors could not be certain which ion was displaced, their data did show evidence of a threshold, which confirms that the 6.1-eV band in fact arises from a lattice defect associated with an atomic displacement process.

Arnold and Compton suggested that an aluminum displacement was energetically more likely, however, subsequent work by Lee and Crawford[10] proved that the 6.1-eV band is actually associated with a lattice defect created by oxygen displacement. Crawford and co-workers have studied the optical properties of sapphire in some detail.[11–15] Lee and Crawford[12] showed that photon excitation at 4.8 eV produced an emission at 3.8 eV. Evans and Stapelbroek[16] showed later that 3.8-eV emission could be produced by both 4.8- and 5.4-eV excitation, and that an absorption band at 6.3 eV was probably also associated with this 3.8-eV emission. They attributed the 3.8-eV emission to the $1B \rightarrow 1A$ transition of an F^+ center, an oxygen vacancy with a single trapped electron.[17]

Pells and Phillips[18] used a somewhat different technique from that of Arnold and Compton[8] to determine a threshold energy for atomic displacement in sapphire. They derived an apparent threshold of 0.40 MeV by observing visible gross lattice damage in a high-voltage electron microscope. They obtained displacement energies of 76 and 18 eV for oxygen and aluminum displacement, respectively.

Displacement damage in sapphire influences its performance as a first-wall insulator in a fusion reactor. In the high radiation environment of a fusion reactor, sapphire must be able to withstand a continuous level of radiation. This ionizing radiation creates electrons and holes by exciting electrons from the valence band into the conduction band. The production of electron-hole pairs leads to a radiation-induced conductivity (RIC) in sapphire. Once in the conduction band, an electron may either recombine with a hole or become trapped at one of a number of defect sites, usually substitutional impurities or point defects. This trapping is the dominant process controlling the RIC.[19] The RIC may be described by the equation

$$\sigma = \sigma_0 + KR^{\delta} , \tag{1}$$

where σ_0 is the conductivity in the absence of radiation or the base conductivity, R is the dose rate, K and δ are constants. The RIC at a given dose reaches a saturation value dependent on temperature and impurity level. Such saturation values have been determined in MgO, Al_2O_3, and $MgAl_2O_4$, under dose and temperature conditions typical of a fusion reactor.[5,19,20]

However, recent studies by Hodgson[6,21] of the RIC in sapphire have revealed that radiation-enhanced electrical breakdown is potentially a serious problem for the use of these materials as insulators in a fusion environment. The RIC reaches saturation as expected but then increases supralinearly, characteristic of the onset of electrical breakdown. This is due to an increase in σ_0 as defined in Eq. (1). Hodgson's work shows that concurrent ionization and displacement damage, in the presence of an electric field, are critical for this phenomenon to occur. Thus, displacement damage plays an important role in the electrical breakdown. The threshold energy for the onset of this displacement damage is therefore an important parameter.

We have previously presented some preliminary data,[22] obtained using time-resolved luminescence spectroscopy, which show that the threshold energy for atomic displacement processes in sapphire is considerably lower than the values determined in the only other studies of this parameter.[8,18] We now present further data on the emission spectra, threshold energies, and electron-trapping kinetics in electron-irradiated α-Al_2O_3 single crystals.

II. EXPERIMENTAL TECHNIQUES

Samples were irradiated by an electron accelerator (Febetron 706, Hewlett Packard, Field Emission Division). The Febetron produces single pulses of electrons of approximately 3-ns full width at half maximum (FWHM) duration, with a maximum energy of 12 J per pulse, and electron energy selectable within the range 0.20–0.60 MeV. The dose per pulse delivered by the electron beam was measured by a "radiachromic" dosimetry technique.[23]

A cylindrically shaped single crystal of α-Al_2O_3 with approximate diameter 2.5 mm and length 18 mm, was obtained from W. and C. Spicer Ltd. (Cheltenham, England). A second crystal was obtained from Union Carbide Corporation (Crystal Products Division, Washougal, Washington). The crystals are referred to hereinafter as "Spicer" and "Union Carbide." Impurity analyses were performed by Element Analysis Corporation (Tallahassee, Florida) using proton-induced x-ray emission, and the results are presented in Table I. Smaller samples were cleaved from each crystal and mounted in an evacuated brass-irradiation cell fitted with suprasil windows perpendicular to the electron beam direction. For liquid-nitrogen temperature experiments, crystal samples were mounted on an aluminum block inside a larger stainless-steel irradiation cell. Quartz windows on either side of the cell enabled the detection system to be aligned perpendicular to the electron beam direction. Conductive cooling of the crystal was achieved using liquid nitro-

TABLE I. Impurity analyses of single-crystal α-Al_2O_3 samples by Element Analysis Corporation (Tallahassee, Florida) using proton-induced x-ray emission (PIXE): (Top) Crystal obtained from W. and C. Spicer Ltd. (Cheltenham, U.K.). (Bottom) Crystal obtained from Union Carbide Corporation (Crystal Products Division, Washougal, WA). The low atomic number elements (C, O, H, N, etc.) represent an unanalyzed component to the sample.

Element	Detection limit 95% conf. (ppm)	Conc. (ppm)	±	Error (ppm)
O		47.076%		0.235%
Al	253.300	52.92%		3.493%
Si	0.142%	0.207%		0.069%
Cl	268.900	552.903		199.0
Ca	79.110	403.866		49.03
Ti	10.480	14.184		4.752
Mn	2.118	1.747		0.871
Fe	1.608	21.941		1.720
Ni	1.084	6.232		0.678
Cu	1.093	9.873		0.887
Zn	1.010	11.809		1.003
Br	2.412	0.856		0.626
Zr	7.195	1.971		1.844
O		47.076%		0.235%
Al	266.300	52.92%		3.493%
Fe	0.478	1.392		0.203
Cu	0.299	0.439		0.113
Zn	0.272	0.316		0.101
Ga	0.254	0.212		0.097

gen, typically to a temperature of ~83 K, as monitored by a copper-constantan thermocouple situated within 2 mm of the crystal.

The luminescence from the samples was focused onto the entrance slit of a monochromator (Spex Industries, Minimate) whose output was monitored by a photomultiplier (EMI, 9783B) and displayed on a fast storage oscilloscope (Tektronix, 7633). Emission intensity-time data were digitized from the oscilloscope using a video camera (Sony, AVC-3250CE) and an LSI-11 computer system. The entire detection system was contained within a double-walled bronze mesh Faraday cage to shield against radio frequency noise generated by the Febetron.

Data showing emission kinetics over the whole time range of detectable emission were obtained by sequentially normalizing consecutive data files. Additional electron-irradiation experiments were performed at Argonne National Laboratory (Argonne, Illinois), using a 3-MeV Van de Graaff and a 19-MeV linear accelerator (LINAC).

III. RESULTS

Figure 1 show the luminescence emission spectra for the Spicer crystal at the indicated delay times after irradiation with 0.48-MeV electrons at room temperature. The emission maximum is observed at 300 nm. We have previously attributed this[22] to the F^+-center emission (an oxygen vacancy with a single trapped electron[17]) due to its proximity to the expected F^+ emission wavelength of ~325 nm (Ref. 15) and its distance from the F emission wavelength of ~415 nm.[15] Subsequent data from the Union Carbide crystal, to be discussed below, where emission is observed at both 300 and 325 nm, suggest otherwise. Initially, in the Spicer crystal, between 144 and 287 ns, the emission decays slowly, however, there is a more rapid decrease in intensity between 287 and 574 ns.

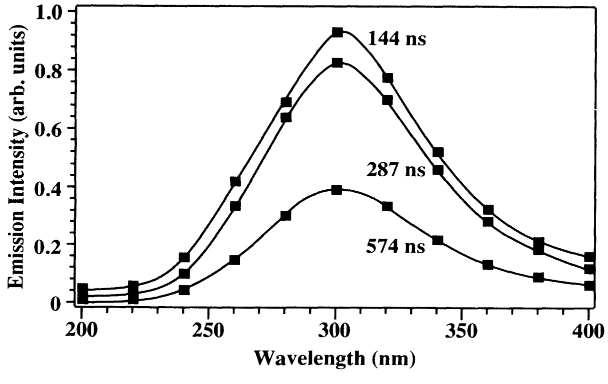

FIG. 1. Luminescence spectra of the Spicer sapphire at the indicated delay times after irradiation by 0.48-MeV electrons in a 3-ns pulse at 293 K.

Luminescence emission spectra for the Spicer sapphire irradiated with 0.48-MeV electrons at 83 K are shown in Fig. 2. While the absolute intensity at 83 K was several times greater than that at 293 K, the general features were similar. The emission maximum was observed at 300 nm, however, the emission bands were somewhat broader than those at room temperature. The rapid decrease in intensity observed between 144 and 287 ns at 293 K was not apparent at 83 K.

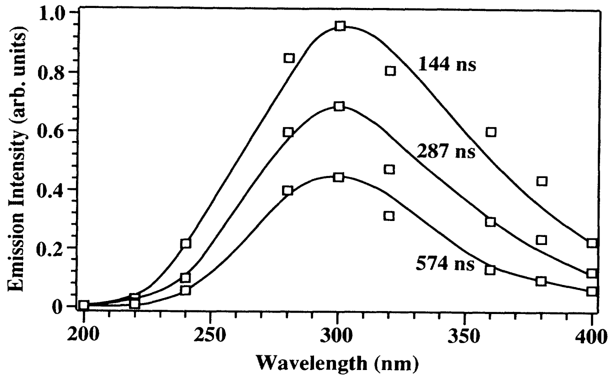

FIG. 2. Luminescence spectra of the Spicer sapphire at the indicated delay times after irradiation by 0.48-MeV electrons in a 3-ns pulse at 83 K.

The threshold energy for atomic displacement processes may be measured by varying the incident electron energy while monitoring a radiation-sensitive property (e.g., conductivity, absorbance, luminescence, etc.). In the present study, luminescence emission is observed and the incident electron energy is progressively decreased in steps, until the threshold is reached, where no emission is observed. From the threshold, the atomic displacement energy, the maximum energy transferred to displaced ions, may be determined.

The results of a threshold determination for the Spicer crystal at 300 nm and 293 K are shown in Fig. 3. A distinct threshold is seen at 0.30 MeV, with an error of less than ±0.01 MeV. Below this energy little or no emission is observed. A threshold determination was also made at 83 K to determine whether there is a temperature dependence for atomic displacement processes in sapphire as has been reported in MgO.[24] Again, a threshold was present at 0.30 MeV. The presence of a threshold confirms that the emission at 300 nm is, in fact, associat-

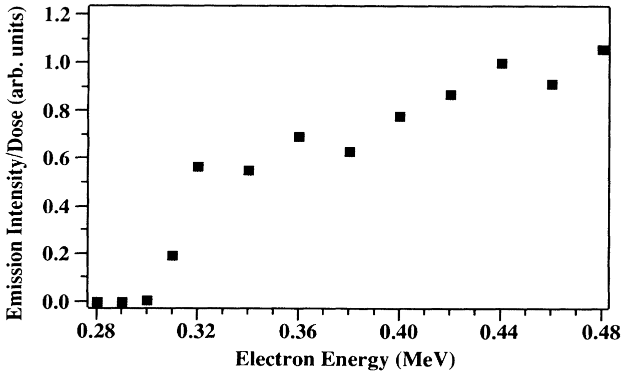

FIG. 3. Emission intensity per unit dose as a function of incident electron energy for the 300-nm emission from the Spicer sapphire at 293 K, 287 ns after the 3-ns electron pulse.

ed with an atomic displacement process and is not due to an ionization process. Such processes would be electron energy independent.

The maximum energy T (in eV) transferable from an incident electron of energy E (in MeV) to a lattice ion of mass number A is given by[17]

$$T=\frac{2147.7E(E+1.022)}{A} . \tag{2}$$

Hence by substituting a threshold energy of 0.30($\pm$0.01) MeV into Eq. (2), an oxygen displacement energy of 53($\pm$2) eV is obtained, in agreement with the displacement energies of oxygen in similar materials.[24,25] If the displaced ion were aluminum, the displacement energy would be 32($\pm$2) eV.

Figure 4 shows the luminescence emission spectrum 287 ns after the electron pulse for the Union Carbide sapphire irradiated at room temperature with 0.48-MeV electrons. In contrast to the Spicer crystal, which showed only one band, occurring at 300 nm, the Union Carbide sample appears to show emission bands at 290–300 nm, and also at about 325 nm, where F^+ emission is expected to occur.[16]

The Union Carbide crystal was also irradiated by 19-MeV (LINAC) and 3-MeV (Van de Graaff) electrons. The spectra are shown in Figs. 5 and 6. The LINAC data were taken 10 ns after the 4-ns electron pulse, and the Van de Graaff data were recorded on a boxcar integrator with a 50-ns gatewidth as a time average between 40 and 90 ns after the 12-ns pulse. Both spectra feature an emission at ~325 nm with a shoulder at ~290–300 nm. The Van de Graaff spectrum was resolved into two Gaussian components, as shown by the solid line in Fig. 6, with band maxima at 298 and 326 nm.

Threshold determinations were made for the Union Carbide crystal at both 300 and 325 nm. It should be noted, however, that because the two bands overlap considerably a threshold determination of either band will contain some contribution from the other. For the 300-nm band, the threshold (Fig. 7) was 0.28 MeV, in contrast to the value of 0.30 MeV in the Spicer sample. The 325-

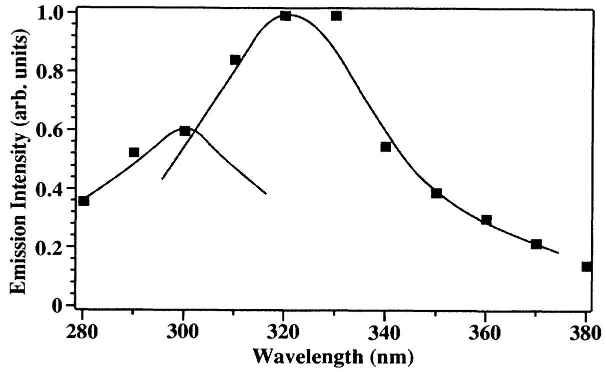

FIG. 5. Luminescence spectrum of the Union Carbide sapphire after irradiation by 19-MeV electrons from a LINAC at room temperature, 10 ns after the 4-ns electron pulse.

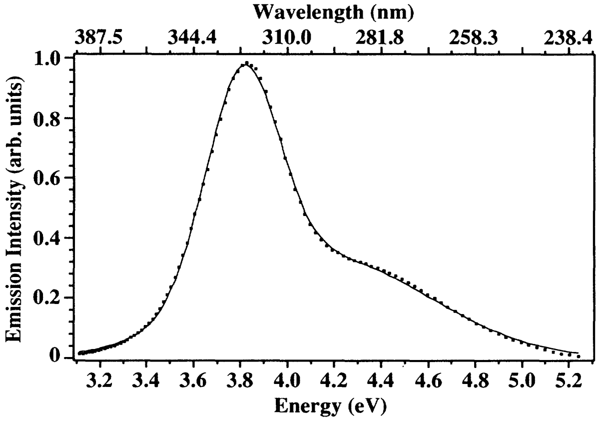

FIG. 6. Luminescence spectrum of the Union Carbide sapphire after irradiation by 3-MeV electrons from a Van de Graaff at room temperature, taken as a 50-ns boxcar integrator gatewidth from 40 to 90 ns after the 12-ns electron pulse. The solid line is a deconvolution of the data into two separate Gaussian components.

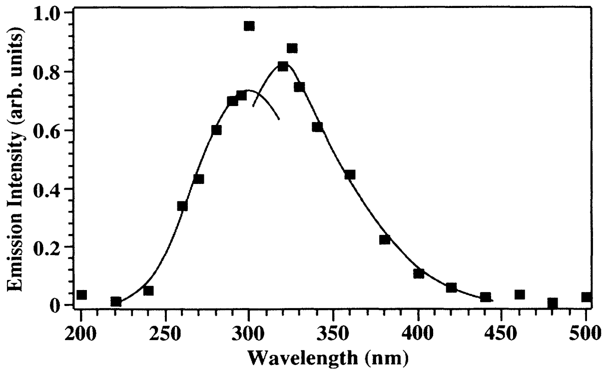

FIG. 4. Luminescence spectrum of the Union Carbide sapphire after irradiation by 0.48-MeV electrons at 293 K, 287 ns after the 3-ns electron pulse.

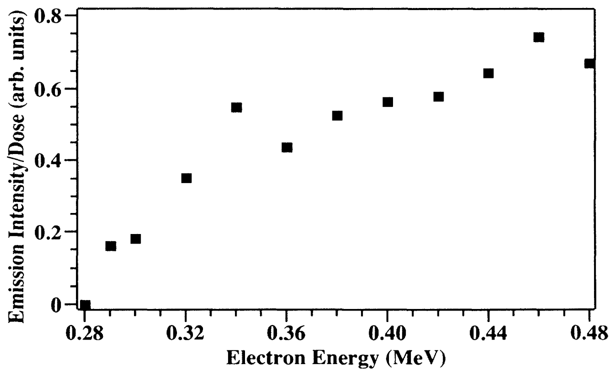

FIG. 7. Emission intensity per unit dose as a function of incident electron energy for the 300-nm emission from the Union Carbide sapphire at 293 K, 287 ns after the 3-ns electron pulse.

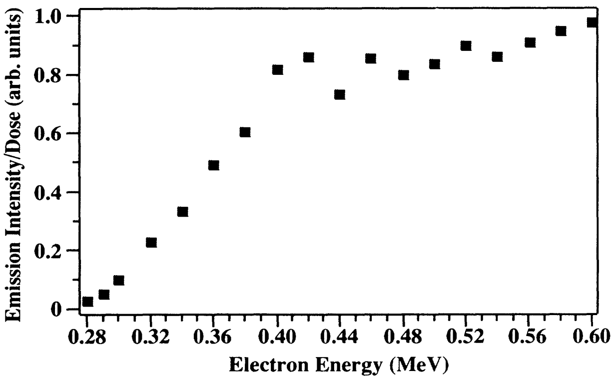

FIG. 8. Emission intensity per unit dose as a function of incident electron energy for the 325-nm emission from the Union Carbide sapphire at 293 K, 287 ns after the 3-ns electron pulse.

nm band also has a threshold at 0.28 MeV, as shown in Fig. 8.

Information about the trapping processes responsible for the RIC may be obtained from analysis of the kinetics of luminescence emission from sapphire. By normalizing consecutive data files from times in the nanosecond region (that is, 10, 20, 50 ns, etc.), to longer times at the detection limits of the experimental apparatus (typically milliseconds to seconds), it is possible to examine the luminescence kinetics for a number of decades of time. Such data were obtained at the emission maximum, 300 nm, for the Spicer sapphire irradiated at room temperature (293 K) with an incident electron energy of 0.48 MeV. The data are shown in Fig. 9. A 0.48-MeV incident electron energy is sufficiently high (given a threshold of 0.30 MeV) to create trapping centers by atomic displacement. The data are presented logarithmically because of the initial rapid decay of luminescence intensity. Several features are apparent. The data span 7 orders of magnitude in intensity and 6 orders or magnitude in time (nanoseconds to tens of milliseconds). There is an initially rapid decay of intensity which subsequently decreases to a plateau level, increases again, and decreases to a second plateau. Such behavior, where the emission shows distinct plateau regions, appears to be a common feature of emission kinetics in similar materials such as CaO and MgO.[26–28] This suggests radiative recombination of electrons and holes, with charge carriers (probably electrons) thermally released from traps at discrete energy levels.

IV. DISCUSSION

Brewer, Jeffries, and Summers[29] proposed a model to describe *F*-center optical behavior in sapphire. The 6.1-eV transition for the observed absorption band is from a ^1s-like ground state to a ^1p-like state lying very close to, or in, the conduction band. The C_2 local symmetry of the *F* center should split the ^1p-like state into three components. However, only one *F* absorption band is observed in contrast to the three F^+ absorption bands.[16] Concentration quenching in neutron-irradiated sapphire, observed in *F*-center emission but not in F^+ emission,[30] provided evidence that the absence of ground-state splitting is due to a greatly extended wave function. Nonradiative decay occurs from the relaxed ^1p-like state to the emitting ^3p-like state. This ^3p-like state, relatively more compact than the ^1p-like state, is split by the C_2 symmetry into $1B$, $2B$, and $2A$ states. Emission at 3.0 eV (413 nm) arises from the two lowest-lying triplet components, which are populated nonradiatively by the ^32A state.

Brewer, Jeffries, and Summers[29] suggested that a radiative decay from the relaxed ^1p-like state to the ground state might be expected to occur at about 4.25 eV (292 nm). Figure 10 shows the Brewer-Jeffries-Summers model with the predicted transition. Such an emission has not been reported by other workers, however, the 298-nm (4.15 eV) band observed in the present study could well arise from such a transition. The observation of similar displacement thresholds (assuming that the considerable spectral overlap does not have a major effect) for the two bands supports the inference that both emissions arise from oxygen displacement processes, one vacancy con-

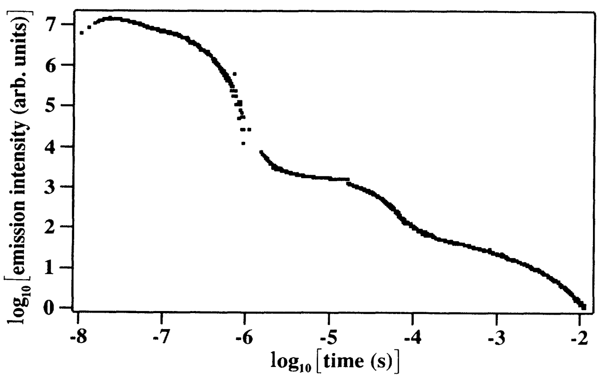

FIG. 9. Emission kinetics for the 300-nm emission from the Spicer sapphire after irradiation by 0.48-MeV electrons in a 3-ns pulse at 293 K.

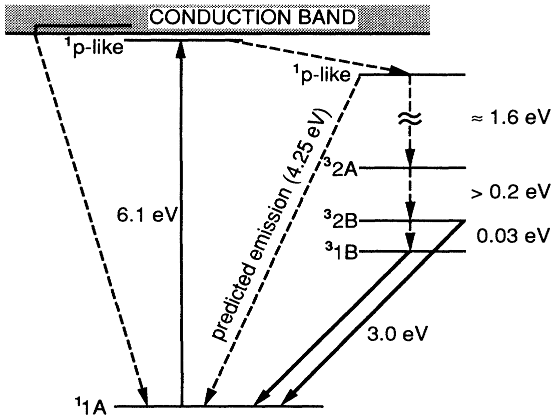

FIG. 10. The model proposed by Brewer, Jeffries, and Summers (Ref. 29) to describe *F*-center optical behavior in sapphire, showing the predicted transition at 4.25 eV.

taining a single trapped electron (the F^+ center), and the other containing two trapped electrons (the F center[17]).

The apparent absence of F-center emission at ~415 nm (3.0 eV) is possibly due to the mode of production of F-type centers. For example, the cathodoluminescence data of Draeger and Summers[31] have shown that the intensity of F emission (at 3.0 eV) relative to F^+ emission (at 3.8 eV) is lower for electron-irradiated than for growth-colored α-Al_2O_3.

As can be seen in Table I, the two crystals vary markedly in impurity content. The Spicer crystal contains substantial amounts of silicon, chlorine, and calcium. The Union Carbide crystal in contrast provides an example of the high-purity levels available nowadays in crystal-growth techniques. Impurities in the Spicer crystal may affect emission properties, but it is unlikely that the very low impurity levels in the Union Carbide crystal would play a significant role in the observed emission.

Despite the extensive research on sapphire, there have been very few experimental determinations of the threshold energy for atomic displacement in this important material. The value determined in the present study, 0.30 MeV, provides the first evidence that earlier determinations may have placed the threshold at a higher energy than is actually the case.[8,18] Given the importance of the atomic displacement process in the subsequent dielectric breakdown in sapphire observed under simulated fusion reactor conditions,[6,21] accurate measurements of this parameter are crucial.

Hodgson[21] referred to the previously measured threshold values of ~0.40 MeV to explain his observation that the increase in base conductivity will not occur when the electron energy is reduced from 1.0 to 0.30 MeV. The present threshold results are fully consistent with this explanation, as an electron beam energy at (or below, in the case of the Union Carbide sapphire) the threshold would not be able to cause atomic displacement.

The experimental data of Arnold and Compton[8] range from 1.77 down to 0.59 MeV. In the present study we conduct observations above, at, and below the threshold energy, thereby directly observing the onset of displacement emission. Arnold and Compton determined the threshold energy by fitting theoretical cross-section curves for atomic displacement to their data, and extrapolating the best-fitting curve (50 eV for aluminum displacement or 90 eV for oxygen) to a threshold energy of 0.43 MeV. In a later study,[9] they made similar measurements, ranging from 1.54 down to about 0.53 MeV, and determining the threshold to be 0.37 MeV (for a 40-eV aluminum displacement or a 70-eV oxygen displacement). The present analysis allows a more accurate determination of the threshold energy. The 83-K data in the present study were inconsistent with the high threshold energy reported by Arnold and Compton[8] for 77-K electron irradiation.

In the case of the threshold determination by Pells and Phillips,[18] it should be noted that visual observation of gross lattice damage in an electron microscope is not a particularly sensitive technique for determining a threshold energy for specific isolated atomic displacement. The dose of electrons (approximately 10^{12} electrons/cm^3) needed to determine threshold values in the present study is some 10 orders of magnitude lower than that required for electron microscopy, and the measured luminescence arises directly from the atomic displacement process itself, as evidenced by the existence of a threshold.

Hughes[32] and Klaffky *et al.*[19] have proposed simple two-trap models to explain the radiation-induced conductivity observed in alumina. These consist essentially of shallow (low-energy) electron traps, and deep (higher-energy) electron traps, with a single hole trap which acts as a recombination center. We have used a similar model in an attempt to explain the kinetic features observed experimentally in electron-irradiated alumina (Fig. 9). This appears to provide both a qualitative and quantitative explanation for the complex kinetic behavior observed in sapphire, and will be presented in greater detail in a subsequent publication.

The trap depths of the two relatively shallow electron traps which empty at room temperature within the time scale of our experiment may be calculated from the observed detrapping times which correspond to the plateau regions. The detrapping time is exponentially dependent on trap depth, as given by the following equation:[33]

$$\tau = s^{-1} \exp\left[\frac{\Delta E}{kT}\right], \tag{3}$$

where τ is the detrapping time in s, s is a frequency factor in s^{-1}, ΔE is the trap depth in eV, k is the Boltzmann constant, and T is temperature in K. Assuming a frequency factor of 10^{12} s^{-1}, a typical value for most materials,[33] the trap depths are 0.36 and 0.48 eV. Klaffky *et al.*[19] calculated trap depths of 0.57 and 0.72 eV from RIC studies on electron-irradiated sapphire from two different manufacturers. Hughes[32] calculated a trap depth of 0.75 eV from the time dependence of x-ray excited electrical conductivity. Cooke *et al.*[34] measured a thermal activation energy of 0.69 eV associated with a 270-K thermoluminescence glow peak, and 0.47 eV for a 210-K peak. Therefore, the trap depths calculated in the present study are similar to those measured in other studies. The role of charge carrier trapping in RIC has been stressed above, so the measurement of these parameters is important in understanding the problem of radiation-enhanced electrical breakdown.

V. SUMMARY

We have measured the luminescence emission spectra and threshold energies for atomic displacement processes in electron-irradiated sapphire single crystals. The Spicer crystal, which contained a significantly higher impurity level than the Union Carbide crystal, features a broad emission band centered at 300 nm. The threshold energy for this band, 0.30±0.01 MeV, corresponds to an oxygen displacement energy of 53±2 eV or an aluminum displacement energy of 32±2 eV.

Analysis of the emission kinetics of the Spicer sapphire at room temperature showed behavior consistent with detrapping from shallow electron traps at 0.36 and 0.48 eV.

In the Union Carbide crystal, two emission bands were observed, at ~298 and ~326 nm. Threshold energies at 293 K for both bands were 0.28±0.01 MeV, corresponding to oxygen displacement energies of 49±2 eV or aluminum displacement energies of 29±2 eV. In view of the consistency with similar crystals[24,25,35] we presume that these thresholds correspond to oxygen displacement.

The emission at ~300 nm observed in both crystals has not previously been reported by other workers. However, it may be due to a transition from a relaxed ^1p-like state to the ground state of the F center, as predicted by Brewer, Jeffries, and Summers.[29] In the absence of other supporting experimental data, the evidence for this assignment is the observation of similar displacement thresholds for the 3.8- and 4.2-eV emissions. As also suggested by Draeger and Summers[31] in their study of α-Al_2O_3, the true nature of F-center luminescence is unresolved, so the present conclusions remain speculative.

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