PHYSICAL REVIEW VOLUME 91, NUMBER 4 AUGUST 15, 1953

Irradiation-Induced Photoconductivity in Magnesium Oxide*

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(Received March 9, 1953)

Crystals of magnesium oxide have been colored by irradiation with ultraviolet light and neutrons. The spectral distribution of optical absorption has the form of a gradually increasing tail extending from the visible region to the far ultraviolet upon which are superimposed several absorption bands. Photoconductivity in single crystals of magnesium oxide was measured by a dc method using a vibrating reed electrometer. The spectral distribution of photoconductivity is characterized by a gradually rising tail with superimposed peaks at 2.1, 3.7, and 4.8 ev, corresponding to known optical absorption bands. A photoconductivity band was found at 1.2 ev which has not been detected by optical absorption measurements. Irradiation of the crystals by ultraviolet light causes an enhancement of the photoconductivity subsequently measured in the 1.2- and 2.1-ev bands. The enhancement effect reaches a saturation level which is independent of the intensity of the ultraviolet light and which is a measure of the density of imperfections in the crystal lattice. The ultraviolet activated region can be displaced by an electric field in such a direction as to indicate that the charge carriers are holes in the valence band. Neutron irradiation of the crystals gives rise to a thermally unstable enhancement of photoconductivity throughout the spectrum and also causes an increase in the level of saturation of the ultraviolet activation. The latter increase is stable at room temperature and indicates that the neutron irradiation produces new lattice defects. This effect saturates with increasing neutron flux. An estimate of the density of lattice defects can be made from the photoconductivity. An energy-level model is proposed to explain the various photoconductivity bands and the enhancement and saturation effects.

INTRODUCTION

SINGLE crystals of magnesium oxide may be colored by irradiation with ultraviolet light,[1] x-rays,[2] neutrons, and high-energy electrons.[3,4] Measurements of the spectral dependence of the optical absorption of the colored crystals have revealed several absorption bands in the visible and ultraviolet regions superimposed upon a gradually rising "tail." Absorption bands corresponding to those produced by irradiation have also been created[5] by the direct addition of excess magnesium and oxygen to the crystal lattice. Previous attempts[2,3,6] to find photoconductivity associated with this optical absorption have been unsuccessful. However, by the refinement of the techniques and the use of more sensitive current detecting apparatus, it has been possible to measure the spectral dependence of photoconductivity in magnesium oxide single crystals. Simultaneous measurements of optical absorption were made by the writer but are not included in this publication since they were in essential agreement with the results of other investigators.[1–4]

EXPERIMENTAL APPARATUS

Most of the samples were plates on the order of one millimeter thick and one centimeter square which had been cleaved from larger crystals obtained from two sources. One group was supplied by the Norton Company[7] and the other group was grown at this laboratory.[8] Aquadag electrodes were painted on opposite faces of the crystals and were air dried at room temperature. The samples were mounted between spring clips on Teflon insulators in an airtight box. Desiccation was found to be essential because of high leakage currents caused by surface moisture. A quartz window was provided in the side of the box to permit the entrance of the irradiating wavelengths. In the visible and near infrared regions, a tungsten filament projection lamp served as the light source, a 100-watt, type H-4 mercury arc was used in the near ultraviolet, and a 1000-watt type H-6 mercury arc was used in the far ultraviolet. After passing through a Gaertner quartz monochromator the radiation was focused by means of a system of front surface mirrors onto the edge of the sample. Provision was made for using a thermopile to monitor the intensity of the light at each wavelength setting. An electric field was placed across the crystal, and the resulting photocurrent was determined by measuring with a vibrating reed electrometer[9] the potential drop which it produced in a high resistance. The highest resistance used was 10^{12} ohms, and currents as low as 10^{-16} ampere could be measured by the constant deflection method.

EXPERIMENTAL RESULTS

1. Characteristics of Photoconductivity

The observed photocurrents were superimposed on a dark current of the order of 10^{-15} ampere which could be balanced out by a bucking voltage in the electrometer. The increase of current produced by illumination was called the photocurrent, and it was found to be a linear function of the intensity of the light and of the

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* Work supported in part by the U. S. Office of Naval Research and a grant from the Radio Corporation of America, Laboratories Division.

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[1] J. H. Hibben, Phys. Rev. **51**, 530 (1937).

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[3] Boyd, Rich and Avery, U. S. Atomic Energy Commission Report MDDC-1508, 1947 (unpublished).

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[6] W. W. Tyler and R. L. Sproull, Phys. Rev. **83**, 548 (1951).

[7] The Norton Company, Niagara Falls 5, New York.

[8] These crystals were grown by H. F. John of this laboratory.

[9] Applied Physics Corporation, Pasadena, California.

electric field strength up to 14 000 volts/cm, the limit of measurement. It was also found that the photocurrent at a given wavelength and light intensity did not depend on whether the light was distributed over the whole edge of the crystal or focused on only a narrow region, as long as the total power received by the sample remained unchanged. When a narrow region in the center of a crystal was illuminated by ultraviolet light, a current was produced which, after an initial decrease by a factor of about two, was constant over a period of many hours. The mechanism whereby constant dc photocurrents can be produced by the illumination of only part of a crystal is not yet understood.[10] Because of the linear dependence of photocurrent upon intensity and electric field strength, the data were reduced to units of induced photoconductivity per unit optical power striking the crystal.

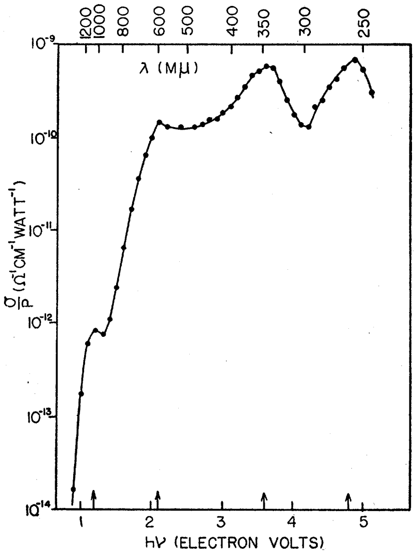

FIG. 1. Completely developed photoconductivity spectrum of a magnesium oxide single crystal, recorded immediately after irradiation by an integrated neutron flux of 10^{17} neutrons/cm^2.

The completely developed photoconductivity spectrum contains several bands which are shown in Fig. 1. The maxima of these bands occur at 1.2, 2.1, 3.7, and 4.8 ev, the latter three corresponding closely to optical absorption bands which were found in the previous investigations. The band at 1.2 ev has not been detected by absorption studies.

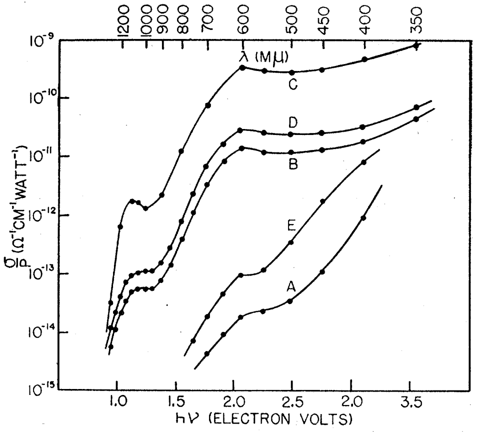

FIG. 2. Photoconductivity spectra of a crystal: (A) untreated, (B) after activation to saturation by 312-mμ light, (C) immediately after neutron irradiation, (D) new saturation level of ultraviolet activation after neutron irradiation and photobleach to curve (E).

2. Ultraviolet Irradiation

Freshly cleaved, untreated crystals exhibited a photoconductivity spectrum similar to curve A of Fig. 2. The gradual rise and the shoulder at 580 mμ or 2.1 ev are characteristic of all photoconductivity spectra observed for magnesium oxide. The 580 mμ band suggests the presence of an energy level situated in the forbidden band so that a transition of 2.1 ev can produce free charge carriers, either in the conduction band or in the valence band. This photoconductivity band may correspond to the optical absorption band which occurs at very nearly the same wavelength and which causes the characteristic visible coloration of the crystals after irradiation. Curve B of Fig. 2 represents the level of photoconductivity in the same crystal after it had been irradiated for eight hours with ultraviolet light at 312 mμ. This treatment produced an increase by a factor of nearly a thousand in the photoconductivity at 2.1 ev and reveals the new band at 1050 mμ or 1.2 ev. The photocurrents in this region had previously been beyond the range of the detecting instruments. The photocurrent at 312 mμ, the wavelength of irradiation, decreased by a factor of two during the first few minutes of the irradiation after which it remained constant. The enhancement of photoconductivity in the visible and infrared regions of the spectrum by irradiation with ultraviolet light and the decrease in the photoconductivity at the wavelength of irradiation correspond closely to the shift in the optical absorption spectrum which can be produced[4] in a similar manner. This effect strongly suggests the transfer of electrons from one type of energy level to another in a manner similar to the process which occurs in alkali halides.[11]

The dependence of the enhancement of photoconduc-

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[10] A series of experiments was carried out to investigate this matter. It seems certain that true dc currents are being observed and that light scattered at the crystal surfaces and within the crystal is not sufficient to cause the observed photocurrents. No increase in photocurrent is observed when the light strikes the electrodes yet an experiment to be described later indicates a distance of travel of the charged particles of only 10^{-3} cm in a field of 1000 volts per cm. It is possible that the ever present dark current, although usually small, may have some bearing on this mechanism.

[11] N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Oxford University Press, London, 1948), 2nd edition, p. 129.

tivity upon the time of irradiation is shown in Fig. 3. The ultraviolet irradiation was interrupted for brief intervals during which the photocurrent produced by light of 580 mμ was recorded. It is apparent that the enhancement had nearly reached saturation after 160 minutes, and with this ultraviolet intensity of 12 μwatts no further increase was observed after three hours of irradiation.

It was also found that if a crystal which previously had been activated by ultraviolet light was irradiated for a long time with yellow light in the 580 mμ band, a decay was produced in the photoconductivity at all wavelengths above 350 mμ. This decay is shown in the right-hand side of Fig. 3, where the photocurrent at 580 mμ is plotted as a function of the time of irradiation by 39 μwatts of light at that wavelength. The rate of decay was greater with more intense light.

Since the enhancement could be removed by irradiation with yellow light, it was possible to make successive activations with ultraviolet. Four different trials were made, each with ultraviolet radiation of different intensity. The saturation level, that is, the maximum level to which 580 mμ photocurrent could be activated, was the same regardless of the ultraviolet intensity. However, the time required to reach saturation was roughly inversely proportional to the intensity. The time required to reach saturation for the various intensities were: 6.25 μwatts, 6 hours; 12 μwatts, 3 hours; 75 μwatts, 35 minutes; and 125 μwatts, less than 30 minutes.

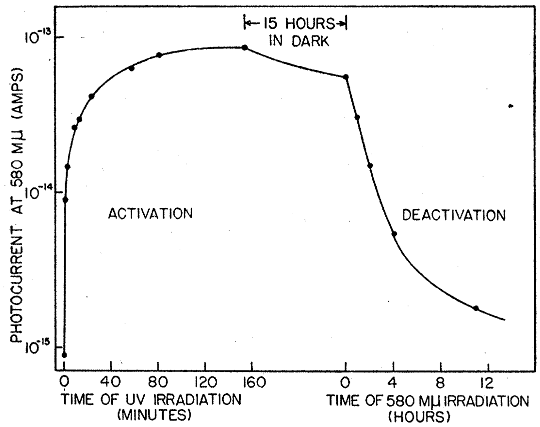

FIG. 3. Time dependence of activation by 12 μwatts of ultraviolet light and of deactivation by 39 μwatts of yellow light. A slight decay occurred overnight in the dark at room temperature.

A decay of the activation similar to that produced by yellow light, but much slower, occurred at room temperature even when the crystal remained in the dark. The curve in the center of Fig. 3 indicates the small decrease which occurred when the sample remained overnight in the dark. The decay in the dark was accelerated at higher temperatures, and the ultraviolet activation disappeared after thirty minutes at 100°C. At higher temperatures, more charge carriers can acquire enough thermal energy to become free of their shallow traps and return to the deeper-lying levels from which they were originally excited by the ultraviolet radiation. It is not likely that 100°C is high enough to cause any appreciable diffusion of lattice defects; hence the decay at this temperature must result only from the transport of electrons.

A crystal which had been activated to saturation by ultraviolet irradiation was heated to about 1000°C in a vacuum better than 10^{-7} millimeter of mercury and cooled slowly to room temperature. After the sample had been vacuum-annealed, it was found that upon reactivation the photocurrent at 580 mμ saturated at a level which was lower by a factor of four than the previous saturation level. The original saturation level had been reproducible after thermal deactivation at low temperatures (20°C to 100°C) and after deactivation produced by irradiation with yellow light, but it could not be reproduced after the high temperature annealing in vacuum. Whereas the decay at low temperature is caused by removing electrons from traps, the lowering of the saturation level is a completely different effect and must be due to a decrease in the number of traps which may be occupied by the electrons. Therefore it is believed that the level of maximum activation is a measure of the density of defects in the magnesium oxide crystal lattice and that the density of these defects can be decreased by annealing the crystal in vacuum. This conclusion is consistent with the previous observation that the level of maximum activation is independent of the ultraviolet intensity.

3. Sign of the Charge Carrier

When the photoconductivity was activated by a well-defined beam of ultraviolet radiation which did not cover the entire crystal, the enhancement was produced in only the localized region which intercepted the beam. It is believed that the mechanism of enhancement is that the ultraviolet photons induce the transfer of charge carriers from one type of energy level to another. During this transfer, the charge carriers pass through an energy band in which they are free to move under the application of an electric field and thereby give rise to the observed photoconductivity. It was found that, by placing a strong electric field across the crystal during the irradiation, it was possible to cause the charge carriers to drift a measurable distance in the direction of the electric field before becoming trapped. This drift was evidenced by a shift of the region of enhancement out of the irradiated area in a direction determined by the sign of the charge carrier. A crystal was illuminated by a beam of light of rectangular cross section and 100 microns in width. The beam passed through the crystal in a direction perpendicular to the applied electric field with the 100-micron dimension parallel to the field. The photocurrent for the wavelength 580 mμ was observed as the light beam was moved along the crystal in the direction of the field. Next, with the beam set in the center of the crystal a 100-micron region was irradiated for one minute, at 335 mμ. This irradiation was sufficient to produce activation but not saturation. Then with 580 mμ light, the beam was again swept across the crystal and the photocurrent was recorded. The results, which are shown in Fig. 4, were found to depend upon the electric field in the crystal at the time of the ultraviolet irradiation. The experiment was performed three times, with no field and with a field of 3800 volts/cm directed to the right and then to the left. Between trials the previous activation was removed by heating the crystal for several hours at 100°C. In each case where a field was applied, the activated region was found to have shifted toward the negative electrode, indicating that the charge carriers are positive, hence they must be holes in the valence band.

Thus the enhancement of photoconductivity at 580 mμ produced by irradiation at 335 mμ is the result of transferring electrons from a level 2.1 ev above the valence band to one 3.7 ev above this band. The mechanism of transfer is probably initiated by the photon excitation of valence band electrons to vacant 3.7-ev levels. Holes which are produced by this transition migrate until they combine with electrons from the populated 2.1-ev levels. During this process, photoconductivity in the 2.1-ev band may increase by a factor of a hundred, whereas photoconductivity in the 3.7-ev band decreases by only a factor of two or three. Therefore there must be a much larger number of 3.7-ev levels to be filled than 2.1-ev levels to be emptied and saturation will occur only when all of the lower-lying levels have become vacant. The 1.2-ev band is also enhanced by 3.7-ev irradiation, increasing simultaneously with the 2.1-ev band and reaching saturation at about the same time. These bands can also be activated by irradiation of energies other than 3.7 ev. By this rather indirect reasoning one concludes that all photoconduction in magnesium oxide is by means of holes, and all the transitions are to and from the valence band.

4. Neutron Irradiation

The crystal discussed in connection with Fig. 2 was irradiated with neutrons in the heavy-water reactor at the Argonne National Laboratory.[12] No attempt was made to control the sample temperature, and it is presumed that it was the same as the ambient pile temperature. After a long irradiation the photoconductivity saturated at the level represented by curve C of Fig. 2, indicating that the maximum level of activation produced by neutrons was a factor of twenty higher than that produced by the ultraviolet irradiation. The sample was left in the dark for several days, during which time the photoconductivity decayed to about the level of curve B. Because of this rather rapid decay in the dark, it is believed that the twenty-fold enhancement produced by neutron irradiation was largely electronic in nature. This activation to a thermally unstable state is referred to arbitrarily as the "primary effect" of neutron irradiation.

A series of irradiations was made in order to determine the rate of activation by neutrons. The neutron-induced photoconductivity approaches a saturation, and it was found that no further increase was produced by an integrated flux of more than 5×10^{15} neutrons/cm^2.

The data of Fig. 1 were taken for a crystal immediately after it had been irradiated by neutrons. The four bands were discussed previously.

After the photoconductivity of the neutron irradiated crystal of Fig. 2 had decayed from curve C to about the level of curve B, the crystal was irradiated with ultraviolet light until saturation occurred. It was found that a new saturation level, curve D, was produced which was higher by a factor of two than the saturation level B observed before the neutron bombardment. If the ultraviolet enhancement saturates only after all the low-lying 2.1-ev energy levels are emptied, as suggested earlier, this new saturation level must result from the production of new lattice defects during the neutron irradiation. As a check on the reproducibility of this effect, this sample was irradiated with 580-mμ light until the photoconductivity had decayed to curve E. Upon subsequent ultraviolet irradiation, saturation occurred again at level D, indicating that the new lattice defects produced by the neutron bombardments are stable at room temperature.

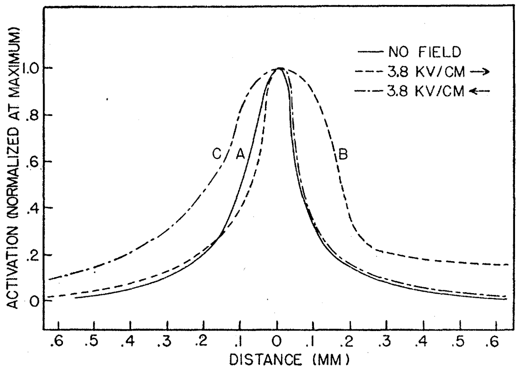

FIG. 4. Displacement of the activated region by an electric field. (A) No applied field, (B) field of 3.8 kv/cm directed to the right, (C) field of 3.8 kv/cm directed to the left.

This sample was later heated for fifteen hours at 100°C and then irradiated with ultraviolet. The saturation level at 580 mμ was still the same as shown by curve D of Fig. 2. After heating the sample for seventy-five minutes at 300°C, the ultraviolet saturation was found to have decreased to the level shown by curve B which represents the condition of the sample before

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[12] Appreciation is expressed to Dr. O. C. Simpson of the Argonne National Laboratory who made these facilities available.

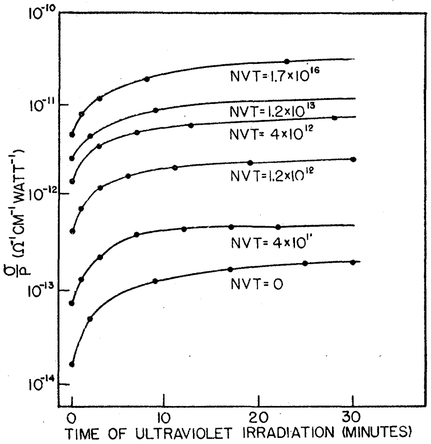

FIG. 5. The enhancement of photoconductivity at 580 mμ by 335 mμ light for various neutron irradiations. *NVT* is the integrated neutron flux in neutrons/cm^2.

neutron irradiation. The crystal was then irradiated by a neutron flux which was larger by a factor of ten than that used previously. The integrated neutron flux in this case was 6.5×10^{16} neutrons/cm^2. The photoconductivity of the sample was at first very high (curve C of Fig. 2), but this "primary effect" could be removed by irradiation with visible light until the sample was in a state near that represented by curve E. Upon subsequent ultraviolet irradiation the photoconductivity measured at 580 mμ saturated at the level shown by curve D, the same as had been obtained after the first neutron bombardment.

The results of the baking procedure and the second neutron irradiation verify the reproducibility of the neutron-produced saturation level D and suggest the following: 1. The level of saturation of the enhancement of photoconductivity by ultraviolet irradiation is stable at room temperature and is a measure of the density of defects in the crystal lattice. 2. This level of saturation can be changed by neutron bombardment in a process which may be called the "secondary effect," as distinguished from the temporary neutron-produced enhancement referred to as the "primary effect." 3. The secondary effect itself reaches a saturation level which is independent of the intensity of the neutron flux and which can be reproduced by further bombardment after the crystal has been thermally annealed. 4. The fact that the ultraviolet saturation level can be lowered by heat treatment after the neutron irradiation indicated that the lattice defects thus produced can be removed by thermal annealing. This indicates that the effect is not due to the introduction of impurities through transmutation. Vacuum annealing also reduced the state of disorder in the crystal since, as was previously mentioned, the ultraviolet saturation of an untreated crystal was lowered by a factor of four by annealing at 1000°C in a vacuum.

The secondary effect was investigated further by making a series of short neutron irradiations on a sample grown at the University of Missouri which initially had a much lower ultraviolet saturation level than the other crystals. This crystal was neutron-irradiated for a fixed time and then bleached to remove the primary enhancement. The crystal was then irradiated with ultraviolet at 335 mμ until a saturation occurred in the photoconductivity at 580 mμ. This procedure was repeated with neutron irradiations of increasing duration, and the results of the experiment are shown in Fig. 5. The curves show that most of the secondary enhancement is produced by an integrated flux *NVT* of 10^{13} neutrons/cm^2, and only a slight further increase is produced by an irradiation one thousand times longer. The saturation of the secondary effect occurred at the same photoconductivity level, $4\times10^{-11}\Omega^{-1}$ cm^{-1} watt^{-1}, for the samples of Fig. 2 and Fig. 5, although before neutron irradiation the saturation levels differed by two orders of magnitude. A third crystal was studied, which before neutron irradiation showed an ultraviolet saturation level the same as the final saturation levels of the other two crystals after neutron irradiation. When it was irradiated with neutrons no secondary enhancement was observed, that is, the ultraviolet saturation level did not change. It would be difficult to present an interpretation of the saturation of the neutron irradiation effect until more data are available regarding its characteristics.

5. Low-Temperature Studies

Although only a very small change is observed in the optical absorption bands of magnesium oxide when its temperature is lowered to that of liquid air, the photoconductivity was found to decrease by a factor of more than 10^5 between 300°K and 90°K in a manner similar to that found for BaO by Tyler and Sproull.[6] The strong dependence of photoconductivity upon temperature suggests that a two-step process may be involved, in which the release of charge carriers depends upon their acquiring enough thermal energy to be released from localized energy levels to which they have been excited by the photons.

DISCUSSION OF RESULTS

1. Photoconductivity Bands

Measurements of the spectral distribution of photoconductivity in magnesium oxide have revealed bands which correspond in wavelength to those which were found in the optical absorption spectra by other investigators. The photoconductivity study has shown that electronic transitions are possible at 1.2, 2.1, 3.7, and 4.8 ev.

The additive coloration experiments of Weber[5] have identified the absorption bands at 2.1, 3.7, and 4.8 ev with excess magnesium and two bands at 4.3 and 5.6 ev with excess oxygen. Since it is believed that neutron bombardment displaces atoms from their normal lattice sites to interstitial positions creating vacancies, both excess magnesium and excess oxygen absorption bands might be produced by the neutron irradiation. The photoconductivity bands which were found after irradiation correspond to the optical absorption bands due to excess magnesium, and bands due to the excess oxygen were not detected. The 4.3-ev oxygen band may have been present but obscured by the two adjacent 3.7- and 4.8-ev magnesium bands. The 5.6-ev band was not observed because equipment limitations prevented measurements at that photon energy.

2. Energy-Level Model

On the basis of the energy transitions represented by the various bands mentioned above, it is evident that localized energy levels exist in the forbidden band. It was shown that the photocurrent is carried by holes in the valence band and that these energy transitions measure the potential of the localized levels above the valence band.

The width of the forbidden band is unknown but is certainly greater than 7.3 ev[13] and, as indicated by x-ray measurements,[14] may be as great as 10 to 15 ev. It is of interest to note that the ionization energy of the O^{--} ions in MgO has been calculated to be 11.8 ev.[15]

Reference should be made to the thermoelectric power measurements of John,[16] which indicate that electrons play the major role in electrical conduction at high temperatures (1300°K). It has also been found[17] that the thermionic work function of magnesium oxide is 2.8 electron volts. The relation of these measurements, both at high temperatures, to the energy-level model is as yet uncertain.

3. Range

In the experiment determining the sign of the charge carrier, the crystal is irradiated in a very narrow region by ultraviolet light. This produces charge carriers which drift, on the average, a small distance down the potential gradient before being trapped. A very rough idea of the range can be obtained by noting this distance of drift. With an electric field of 3.8×10^3 volts/cm the displacement of the region of activation can be estimated from Fig. 4 to be about 5×10^{-3} cm. Most of the photoconductivity experiments were performed with a field of 1 kv/cm, in which case the range would be on the order of 10^{-3} cm.

4. Quantum Efficiency

If n charge carriers are created and trapped in the time t, each moving a distance x before being trapped, the photoconductivity current indicated by an external detector for a sample of length L is

$$i=nex/tL. \tag{1}$$

If radiation of frequency ν falls on the crystal for the time t, the number of photons absorbed is

$$N=Pt/h\nu \tag{2}$$

where P is the optical power absorbed in the crystal. Solving Eq. (1) for n and writing the fraction n/N, one obtains the quantum efficiency,

$$n/N=Lih\nu/xeP, \tag{3}$$

independent of any assumption of the lifetime. Substitution of values for a typical sample shows that n/N increases from 10^{-7} electron/photon at 1050 mμ to 10^{-4} electron per photon at 255 mμ. These efficiencies are small but are not unreasonable considering the extremely low conductivity of magnesium oxide.

5. Density of the Color Centers

Using the value 10^{-4} electron/photon for the quantum efficiency at the wavelength of the ultraviolet irradiation and knowing the time required to saturate the photoconductivity at 580 mμ, one can make an estimate of the number of electrons required to fill all of the 2.1-ev levels. The number of electrons released during the time T required to produce this saturation is obtained from Eq. (1) by substituting $t=T$.

$$\eta=\rho V=(n/N)(PT/h\nu), \tag{4}$$

where ρ is the density of centers and V is the volume of the crystal irradiated. From this calculation one finds there are about 2×10^{14} centers/cm^3. This calculation was carried out for the crystal of Fig. 2 following neutron irradiation and represents the saturation density of 2.1-ev centers. Since the 1.2-ev band saturated with about the same irradiation as required for the 2.1-ev band, approximately the same density of centers must be associated with levels of that energy.

In terms of other physical quantities, the range per unit field strength,

$$x/F\propto l/\rho, \tag{5}$$

where l is the mean free path for collisions with the lattice and ρ is the density of trapping centers. It has been shown[18] that in KCl containing 10^{16} centers/cm^3 the range per unit field strength, x/F, was about 2×10^{-8} cm^2/volt. In MgO, x/F is about 10^{-6} cm^2/volt, indicating a density of defects on the order of 2×10^{14} centers/cm^3, if the mean free path is about the same in both materials. The close agreement of the results of the two calculations is certainly fortuitous, but it is believed that the order of magnitude may be significant.

The author is indebted to Professor A. S. Eisenstein for many discussions concerning this investigation and his helpful criticism of the manuscript.

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[15] J. Yamashita and M. Kojima, J. Phys. Soc. Japan 7, 261 (1952).

[16] H. F. John (private communication).

[17] J. R. Stevenson (private communication).

[18] Reference 11, p. 127.