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**RADIATION AND CHARGE INJECTION IN Al_2O_3
USING NEW TECHNIQUES**

Richard J. Powell

RCA LABORATORIES
Princeton, New Jersey 08540

17 JULY 1975

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trap-assisted tunneling mechanism known to be active in Al_2O_3 . Interpretation of the results is complicated by the presence of both electron and hole traps. Nevertheless, two limiting models are proposed to provide a qualitative explanation.



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PREFACE

This Scientific Report describes work done at RCA Laboratories, Princeton, NJ, under Contract No. F19628-74-C-0132 in the Integrated Circuit Technology Center, J. H. Scott, Director. The Project Supervisor is K. H. Zaininger and the Project Scientist is R. J. Powell. The Air Force Project Monitor is Sven A. Roosild.

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I. INTRODUCTION

The electronic processes in MOS gate insulators have recently been the subject of considerable research effort [1-7]. To a large extent such effort has been directed toward understanding the mechanisms of radiation-induced space charge accumulation. Studies [6,7] of SiO_2 -MOS structures with bandgap light have revealed new information regarding the electronic processes in SiO_2 . In particular, it has been shown that under positive gate bias holes transport to the vicinity of the Si- SiO_2 interface, where a significant number become trapped. The trapped charge enhances the interface field, resulting in electron tunneling currents that can be much larger than the photocurrents. These experiments have been extended to Al_2O_3 in an effort to provide further understanding of the electronic processes involved in this material. In this report we describe the results and give an interpretation of vacuum UV experiments on pyrolytic Al_2O_3 gate insulators.

1. J. F. Verwey, J. Appl. Phys. 43, 2273 (1972), and Appl. Phys. Letters 21, 417 (1972).
2. E. Harari and B. S. H. Royce, IEEE Trans. Nuclear Science NS-20, 280 (1973).
3. R. J. Powell and G. W. Hughes, IEEE Trans. Nuclear Science NS-21, 179 (1974).
4. O. L. Curtis, J. R. Srour, and K. Y. Chin, J. Appl. Phys. 45, 4506 (1974).
5. R. C. Hughes, J. Chem. Phys. 55, 5442 (1971), and Phys. Rev. Letters 30, 1333 (1973).
6. R. J. Powell and G. F. Derbenwick, IEEE Trans. Nuclear Science NS-18, 99 (1971).
7. R. J. Powell, Paper (#R 9526) to be published in J. Appl. Phys., October 1975.

II. EXPERIMENTAL DETAILS

The samples used in the experiments described in this report were prepared by pyrolytic deposition of Al_2O_3 at 975°C on $10\ \Omega\text{-cm}$ n-type $\langle 100 \rangle$ silicon. Semitransparent aluminum-gate electrodes, 1 mm in diameter, were evaporated to produce a sheet resistance of about $20\ \Omega$ per square. Since the absorption depth in Al_2O_3 for 10.2-eV light is approximately $150\ \text{\AA}$ [8], an Al_2O_3 thickness of $2000\ \text{\AA}$ was used to prevent light from reaching the Si- Al_2O_3 interface. This precaution was taken to prevent any contribution to measured photocurrents by electron photoinjection from the silicon into the Al_2O_3 .

The apparatus used in these experiments is illustrated schematically in Fig. 1. The sample is enclosed in a small vacuum chamber coupled directly to a vacuum monochromator. Light from the monochromator is directed onto the

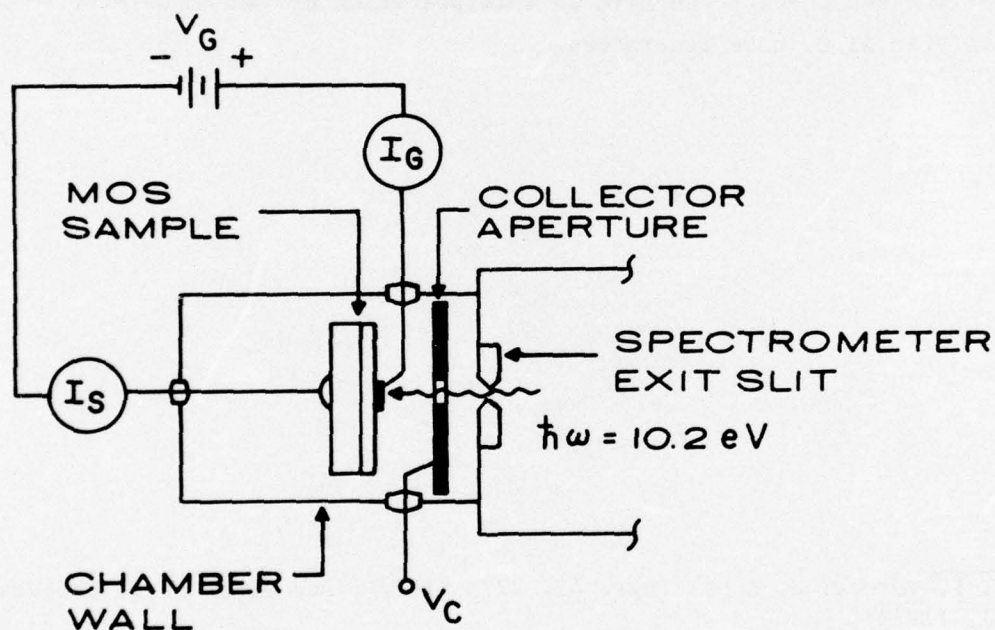


Figure 1. Experimental apparatus used for radiation-induced current measurements.

8. E. T. Arakawa and M. W. Williams, *J. Phys. Chem. Solids* **29**, 735 (1968).

semitransparent gate electrode of the MOS sample through a collector-aperture, which functions to confine the light to the gate electrode and to measure the magnitude of emission into vacuum from the electrode. Two electrometers were used to monitor both the gate current, I_G , and the substrate current, I_S .

The effect of emission current was assessed by grounding the positive battery terminal to the chamber wall and observing the difference between I_G and I_S as the collector voltage was varied from -2 kV to +2 kV. With $V_c = 2$ kV, I_G was slightly less (about 15%) than I_S owing to the electron emission from the gate. With $V_c = -2$ kV, I_G and I_S measured alike. The emission current was measured in the collector lead and found to be less than 15% of the sample photocurrents. A complete experiment was done with the connections as shown in Fig. 1 and repeated with the positive battery terminal grounded to the chamber and with $V_c = -1$ kV. No differences could be observed. With the connections shown, any contribution to I_G and I_S from vacuum photoemission requires that an electron be emitted from the substrate and find its way to the positively biased gate. The likelihood of either process occurring is extremely remote. The precautions described above were taken to ensure that the measured photocurrent results from transport through the oxide film.

From the bias polarity shown in Fig. 1, it is concluded that the measured photocurrent results from electron-hole pairs generated in the absorbing region in the Al_2O_3 near the gate electrode. The absolute light intensity incident on the electrode area was measured by shining the beam through the electrode evaporation mask onto a calibrated cesium-telluride photodiode.* The fraction of incident intensity absorbed in the Al_2O_3 film was calculated from the standard equations for an absorbing film on an absorbing substrate [9], using the optical constants of Al_2O_3 [8] and aluminum** at 10.2 eV.

9. G. Hass and L. Hadley, in *American Institute of Physics Handbook*, Second Edition (McGraw-Hill Book Co., New York, 1963), pp. 6-104,-105.

*The photodiode was calibrated by comparison with a secondary standard traceable to NBS.

**J. G. Endriz, RCA Laboratories, Princeton, NJ, private communication.

III. EXPERIMENTAL OBSERVATIONS AND INTERPRETATION

A. THE TIME DEPENDENCE OF RADIATION-INDUCED CURRENTS

Using the apparatus described in Fig. 1, we measured the time and gate voltage dependence of currents flowing through the aluminum oxide structure. The results are summarized in Fig. 2, which illustrates the time dependence of current for different applied voltages with positive gate bias. The first observation to be made is that the initial current, i.e., the current which flows when the radiation is first initiated, varies significantly with the applied field, but that even for the field of 3 MV/cm the current is far below the line marked 100% Q.E. in the illustration. The 100% quantum efficiency

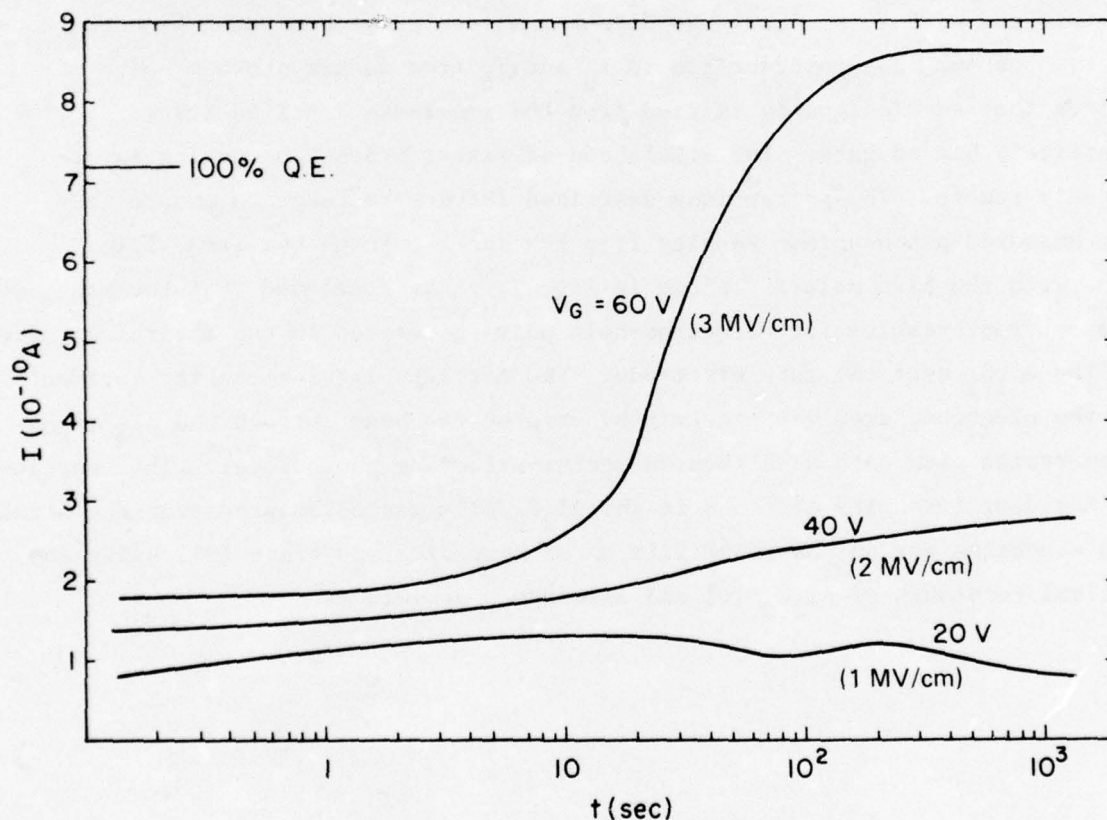


Figure 2. Time dependence of current during irradiation of a 2000-Å Al_2O_3 film with different applied fields under positive gate bias. The photon energy was 10.2 eV, and the photon flux was approximately $5 \times 10^{11} \text{ cm}^{-2} \text{ s}^{-1}$.

(Q.E.) line corresponds to the current one would measure if there were a generation of one electron-hole pair per photon absorbed in the Al_2O_3 and if the total charge transported across the oxide were one electronic charge per absorbed photon. The fact that this photocurrent is less than the 100% Q.E. line indicates that either the net generation of electron-hole pairs is significantly less than unity or the charge transported per absorbed photon is less than one electronic charge. This is discussed in more detail below.

The major features of the current vs time curves shown in Fig. 2 are quite similar to those that have been observed and reported for silicon dioxide [7]. For small applied fields the current is initially almost constant, with some minor variations, and finally begins to decay with time at a rather steady rate. This decay is interpreted to be due to the trapping of some holes near the gate electrode, resulting in a reduction of the field in the light-absorbing region, as previously suggested [7] in the case of SiO_2 . When the gate voltage is increased to 40 V, which corresponds to 2 MV/cm in the Al_2O_3 film, the current is seen to increase monotonically. This current enhancement phenomenon is believed to be due to the tunneling of electrons from the silicon into the Al_2O_3 ; this tunneling results from the increased field as positive space charge accumulates in either the bulk or the interfacial region near the silicon. Finally, for a gate voltage of 60 V, which corresponds to 3-MV/cm applied field, the current rises very rapidly with time, owing to the same mechanism. In this case, the combination of the trapped positive space charge in the film and the applied field of 3 MV/cm is sufficient to produce rather large tunnel injection currents that result in the rapidly increasing total current.

The interpretation of the current as being a combination of tunnel injection currents from the silicon electrode and the photocurrent generated by the vacuum ultraviolet light may be supported by the following piece of evidence: If the incident VUV light is turned off after a short time, the current drops immediately to zero, corresponding to the expected behavior of a photocurrent. However, if after sufficient time for the current enhancement effect to be observed (e.g., 10 s for the 60-V curve in Fig. 2), the light is turned off, there is an instantaneous drop in the current approximately equal to the initial photocurrent; this is followed by a slowly decaying dark current. A set of typical current characteristics as a function of time, with interruption of the

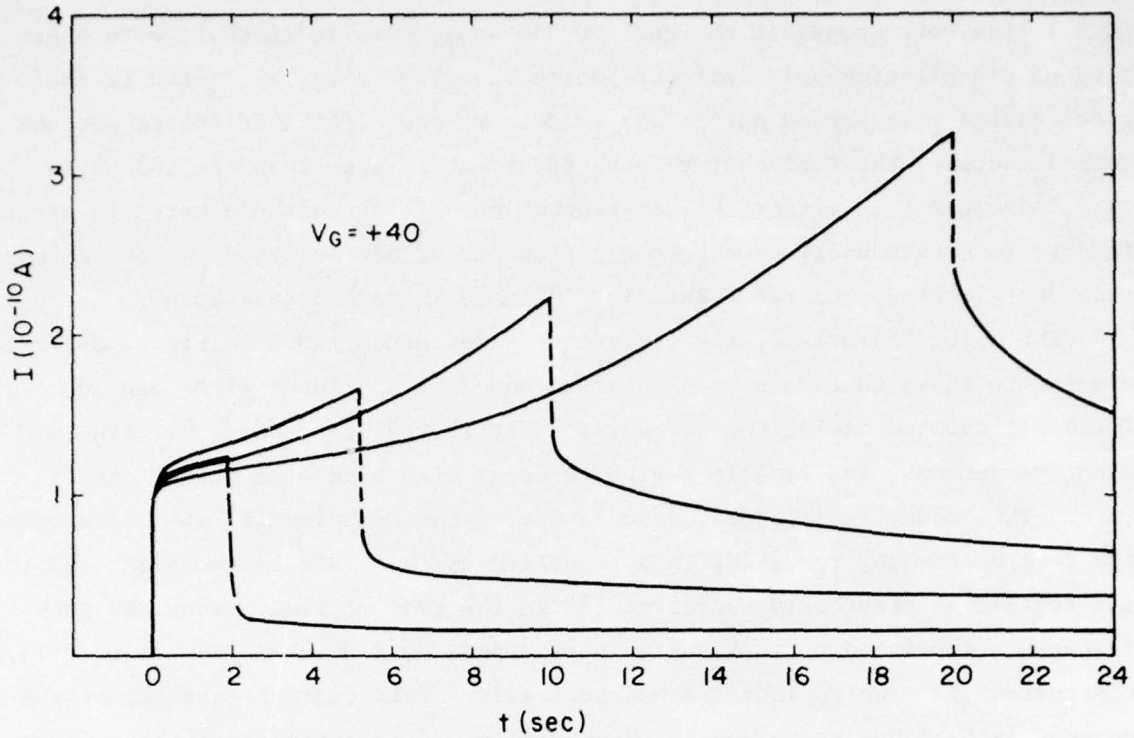


Figure 3. Time dependence of current between initiation and cessation of radiation. Irradiation started at $t = 0$ for each sample. Turn-off is indicated by the dashed near-vertical portion of each curve.

light, is depicted in Fig. 3. In each case the light is turned on at $t = 0$. The light remains on as the current begins to rise due to the enhancement effect and then is turned off at a different time for each curve. The abrupt drop in the current corresponds to the cessation of the VUV light. The variations in the initial currents and also in the rates of current increase among the different curves shown occur because each curve corresponds to a different MOS device on the same wafer, and these differences are believed to be due to sample variations. The important observation to be made regarding these curves is that the current enhancement effect begins immediately, and that, upon cessation of the light, the instantaneous drop in current is caused by the elimination of the photocurrent that has been flowing. The remaining current is that due to electron injection from the silicon electrode. It is noted that, in every case, termination of the light results in an instantaneous drop in the current by an amount approximately equal to the initial photocurrent, followed

positive charge trapped in the oxide. It is not known whether holes are mobile or immobile in Al_2O_3 . Therefore, we have shown two possible cases in Figs. 4(c) and 4(d), corresponding to mobile and immobile holes. In either case, there is an accumulation of positive space charge in the oxide, which eventually will increase the field near the silicon electrode until electron injection begins. This electron injection by either direct tunneling or tunneling into the conduction band will begin to annihilate trapped holes. Eventually, an equilibrium will be reached when a detailed balance occurs between the number of injected electrons that annihilate holes and the number of holes that are generated by the vacuum ultraviolet radiation. Because of the very strong exponential dependence of the tunnel-injection mechanism on the applied field [3], the equilibrium in this case will be reached with the interface field approximately equal to the field at which significant injection begins (~ 1.5 to 1.8 MV/cm). This may be verified experimentally by observing the C-V characteristics before and after irradiation, as shown in Fig. 5. Note that as expected the initial and after-bias application C-V curves are identical, since the bias application does not produce the injection of electrons by the high-field injection mechanism. The post-irradiation C-V curve, however, shows a negative flatband shift of approximately 12.5 V. Since the flatband voltage shift of an MOS capacitor is a measure of the electric field at the Si- Al_2O_3 interface when the applied field is zero, the interface field during irradiation can be estimated from

$$|\xi(0)| = |(V_G - \Delta V_{\text{FB}})/d| \quad (1)$$

where $\xi(0)$ is the interface electric field, V_G is the applied gate voltage, ΔV_{FB} is the flatband voltage shift, and d is the oxide thickness. In this work the magnitude of ΔV_{FB} was measured immediately following radiation using the C-V technique. In Al_2O_3 , ΔV_{FB} tends to shift slowly in the positive direction upon cessation of the radiation. Hence, the field will be underestimated by using the measured ΔV_{FB} in Eq. (1). Nevertheless, if one calculates the magnitude of the interface field for the case shown in Fig. 5, using the observed ΔV_{FB} of 12.5 V and the gate voltage of 20 V, he calculates an interface field of 1.6 MV/cm. This interface field is quite close to the value for which it is typically observed that large electron injection effects occur [3].

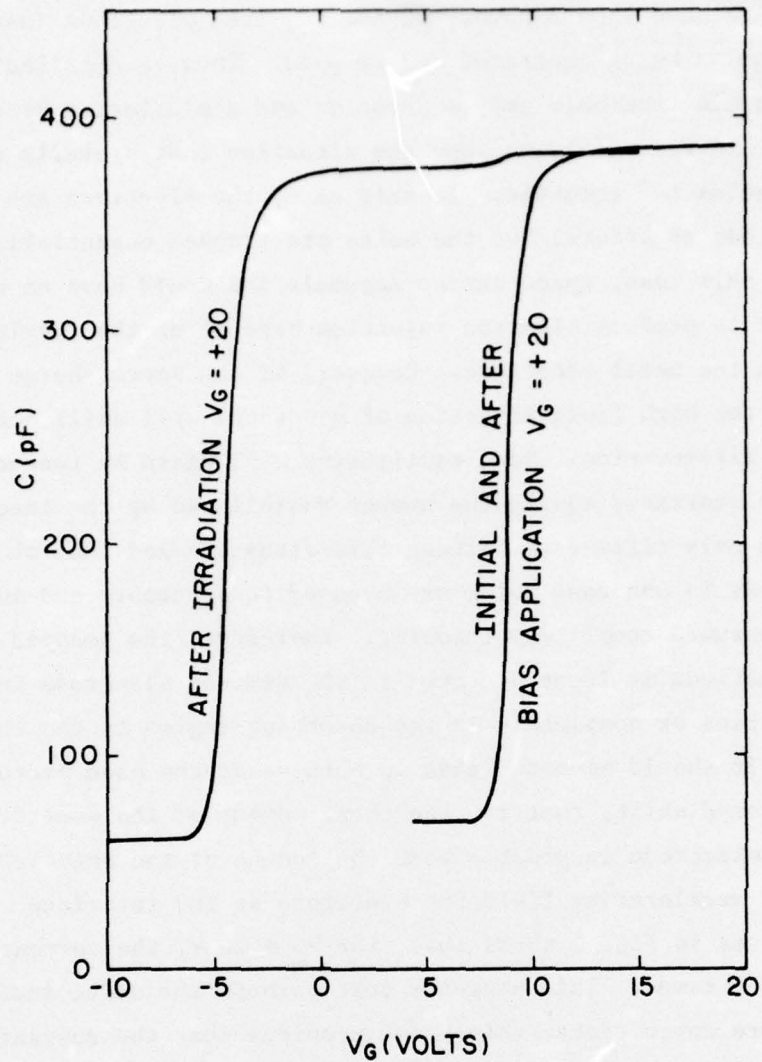


Figure 5. Capacitance-voltage curves of an Al_2O_3 MOS device before and after 20-V bias application, and following irradiation with $V_G = +20$ V. The small kink in the post-irradiation curve near $V_G = 8$ V is due to the small region of the electrode which was covered by the probe and therefore shielded from VUV exposure.

In Figs. 4(c) and 4(d) we show the equilibrium situation during irradiation for the two cases. In Fig. 4(c) we have the case for mobile holes in which the electron-hole pairs are generated near the outer extremities of the oxide, the electrons are swept out to the gate electrode, and the holes transport and trap somewhere in the vicinity of the silicon interface. In this case

equilibrium is reached when a number of the injected electrons just annihilates the number of holes being generated and trapped. Thus, a detailed balance is reached between electron-hole pair separation and annihilation by the incoming electron flux. In Fig. 4(d), we show the situation that prevails during equilibrium, when holes are immobile. In this case, the electrons are swept out to the gate electrode as before, but the holes are trapped essentially where generated. In this case, space charge accumulation would have to be significantly greater to produce electron injection because of the imaging of that space charge in the metal electrode. However, if the space charge accumulation is sufficient, the high field injection of electrons will still occur as indicated in the illustration. Now, equilibrium will again be reached when the number of holes generated equals the number annihilated by the incoming electron flux. The only difference between this situation and that shown in Fig. 4(c) is that in one case holes are assumed to transport and in the other case they are assumed completely immobile. Therefore, the trapped space charge in the two situations is located nearer to the silicon electrode in the case of transportable holes or completely in the absorbing region in the case of immobile holes. It should be noted that in both cases the band picture indicates a negative flatband shift; that is, the total moment of the positive charge about the gate electrode is greater than the moment of the negative charge, providing a net accelerating field for electrons at the interface. Re-examination of the curves in Fig. 2 shows that, for $V_G = 20$ V, the current is still decaying for long times. This suggests that perhaps the model indicated in Fig. 4(d) is more appropriate; this model predicts that the current will decay with time as the space charge accumulates in the absorbing region. In this case, equilibrium will be reached when the photocurrent has decayed to a rather small value and the number of holes being trapped is just balanced by those being annihilated by the incoming electron flux. Note that the model of Fig. 4(c) would not predict a decaying photocurrent since there is little effect on the field in the absorbing region if most of the charge is trapped near silicon interface. However, it is too early to tell from the data shown which model is more applicable, and it is necessary to do some measurements in which the actual ac photocurrent is measured to determine how the photocurrent component itself varies with time.

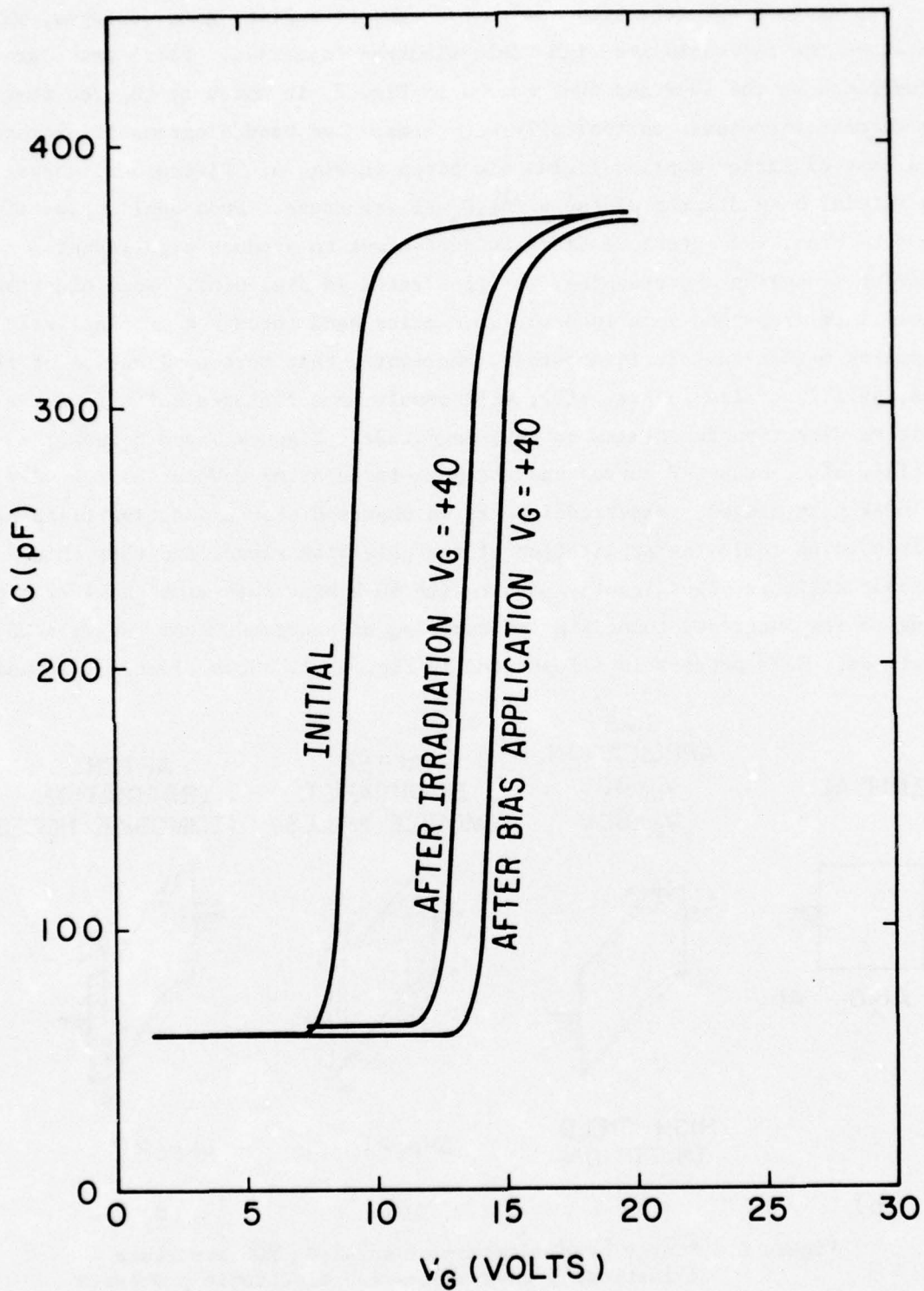


Figure 7. Capacitance-voltage curves of an Al₂O₃ MOS structure before and after 40-V bias application and following irradiation with V_G = +40 V.

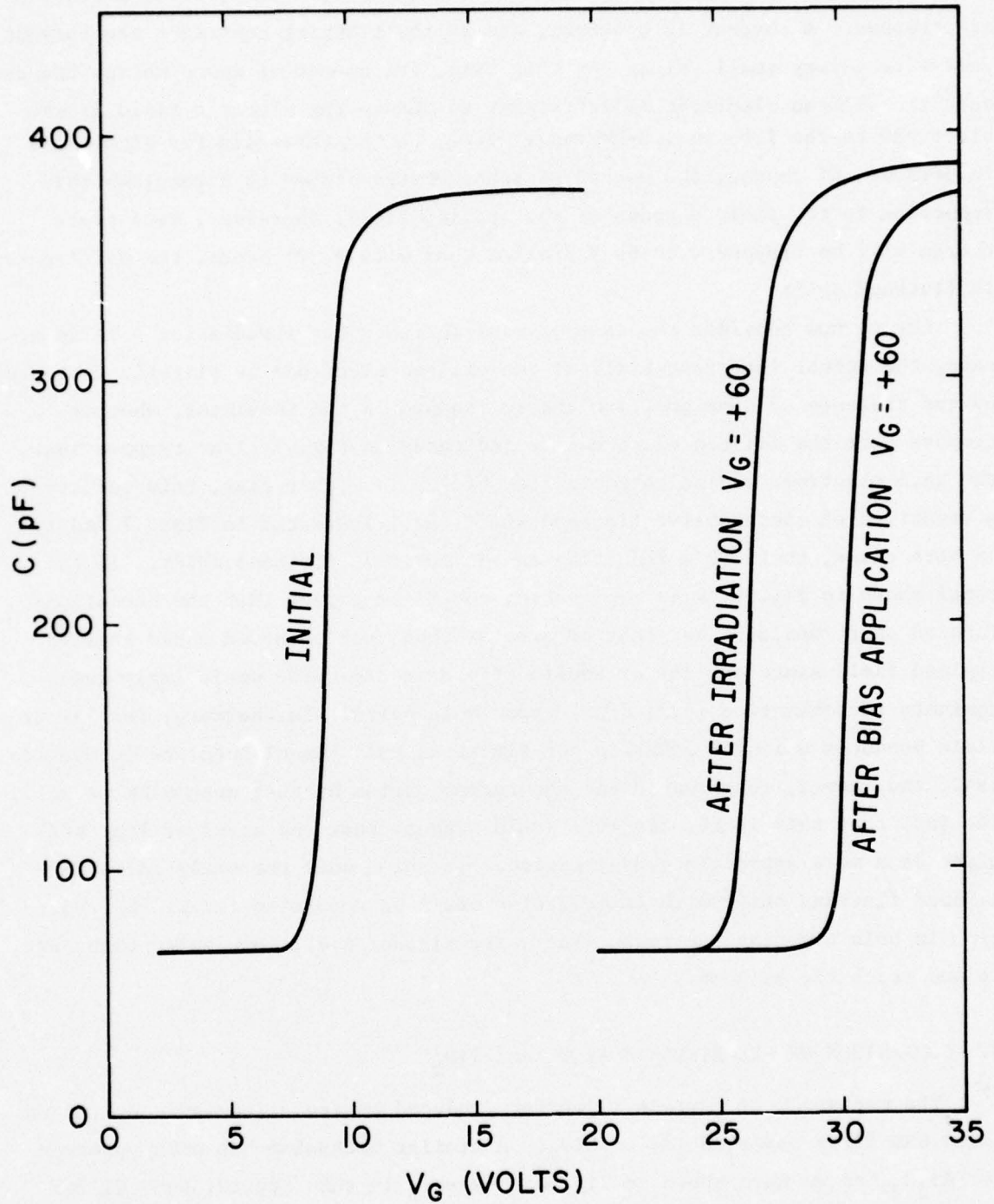


Figure 8. Capacitance-voltage curves of an Al₂O₃ MOS structure before and after 60-V bias application and following irradiation with V_G = +60 V.

directly into traps and some tunnel into the conduction band, where a fraction are trapped. A current is observed, and as the trapping continues the current decays to a very small value. At this time, the amount of space charge trapped near the silicon electrode is sufficient to reduce the electric field at the electrode to the 1.6- to 1.8-MV range, i.e., to the threshold for high-field injection. Of course, the amount of space charge needed to accomplish this reduction in the field depends on the applied field; therefore, more space charge will be trapped with 60 V applied than with 40 V; hence, the differences in flatband shift.

Let us now consider the case of equilibrium under irradiation. In this case, the actual interface field at the silicon electrode is slightly increased by the presence of some positive charge trapped in the insulator, whether trapped near the silicon electrode as indicated in Fig. 6(c) or trapped near the gate electrode as indicated in Fig. 6(d). In either case, this results in a reduction of the positive flatband shift, as illustrated in Figs. 7 and 8. In both cases, there is a reduction in the positive flatband shift. If the model shown in Fig. 6(d) is applicable, one might expect that the radiation-induced shift would be as great or greater than that observed under smaller applied field since the larger applied field in the oxide would imply less geminate recombination [5,7] of electron-hole pairs. Furthermore, the larger field produces a longer *Schubweg* for electrons that tunnel into the conduction band, and, hence, there would be less recombination by that mechanism as well. The fact that this is not observed would suggest that the model of Fig. 6(c) might be a more appropriate description. In this model the small radiation-induced flatband shift with large fields could be accounted for by the reduction in hole trapping due to the increased fields; i.e., more holes transport to and reach the silicon.

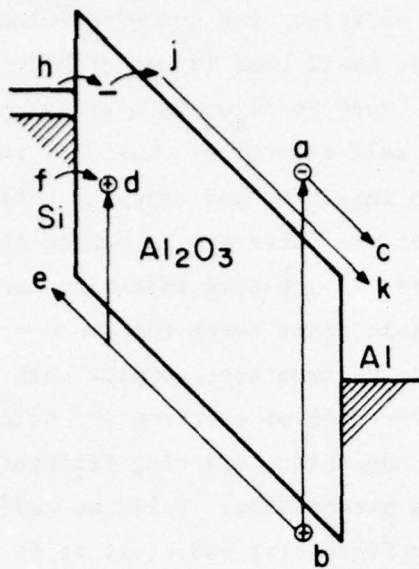
C. COMPARISON OF PROCESSES IN Al_2O_3 and SiO_2

The mechanism of current injection produced by radiation-induced trapped holes was first observed [7] in SiO_2 . A similar mechanism has been observed for Al_2O_3 , as we have shown in Fig. 2; however, in this regard, some differences between SiO_2 and Al_2O_3 should be noted. First, in SiO_2 high field electron injection requires interface fields of the order of 6 to 7 MV/cm, the region of significant Fowler-Nordheim tunneling. In Al_2O_3 this injection be-

gins for fields of the order of 1.5 MV/cm. In addition, the number of electron traps in high-quality SiO_2 films typically is so small that it cannot be measured. In contrast, the number of electron traps in Al_2O_3 is very large; therefore, the injection mechanism in Al_2O_3 is self-quenching. That is, the application of a high field results in electron injection and trapping until the trapped negative charge reduces the field at the interface to quench the electron injection. It is apparent that the presence of this injection mechanism and the presence of electron as well as hole traps makes the processes involved in the radiation-induced charging in Al_2O_3 much more complex than that in SiO_2 . Although it is clear that the processes of electron and hole trapping are competitive in the resulting flatband shift following irradiation, it is not clear just what electronic mechanisms predominate. While we have shown two rather distinctly different models in Figs. 6(c) and 6(d), it is also quite conceivable, and perhaps more reasonable, that an intermediate case actually exists; for example, one in which hole traps exist throughout the oxide and holes have a finite probability of transport. In such a case, the band picture would be intermediate between the two cases illustrated. We have presented arguments which favor the model shown in Fig. 6(c); however, this model would predict a constant photocurrent component since the field in the absorbing region is unaffected by the trapped charge. In fact, observation of Fig. 3 shows that the photocurrent component as indicated by the dashed lines appears to decrease with time. Comparison of the initial photocurrent at $t = 0$ and a photocurrent at 20 seconds shows this effect.

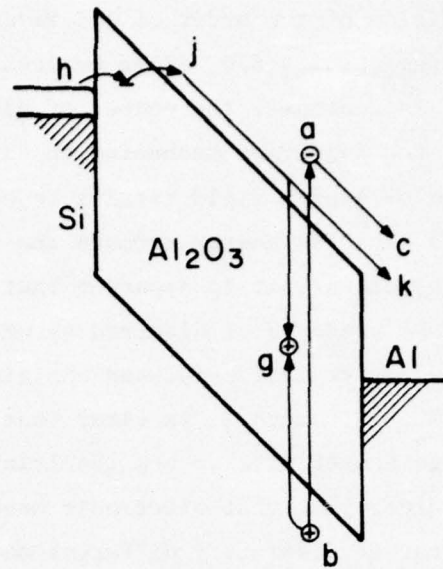
D. THE STEADY-STATE PHOTOCURRENT

Next, we consider two models to explain the steady-state component of photocurrent in Al_2O_3 under irradiation. As we have noted previously, photocurrent components of the currents in Fig. 2 are much smaller than the maximum theoretical value that would obtain if each absorbed photon produced exactly one electronic charge traversing the oxide, i.e., the 100% Q.E. line in Fig. 2. There are at least two possible ways in which this reduced effective quantum efficiency can be explained. Let us consider first the case in which holes are mobile; a model that illustrates the electronic processes is given in Fig. 9(a). Here we designate the electron-hole pair creation by process a-b. Following this electron excitation, the electron is swept out to the aluminum electrode



MOBILE HOLES
INTERFACE LOCALIZED TRAPS

(a)



IMMOBILE HOLES
HOLES TRAP IN BULK WHERE
GENERATED

(b)

Figure 9. Illustration of two possible models to explain the reduced quantum efficiency for photocurrents in Al_2O_3 MOS structures.

as denoted by process a-c, and the hole is swept out to the silicon electrode, marked process b-e or process b-d followed by f-d, in which a tunneling electron annihilates the trapped hole. The electron in traveling to the aluminum electrode contributes to the external circuit a fraction of an electronic charge given by the distance a-c divided by the oxide thickness. Similarly, the hole transporting to the silicon electrode contributes to the external circuit a fraction of an electronic charge given by distance b-e divided by the oxide thickness. Clearly then, the combination of the electron and hole transporting to their respective electrodes contributes one electronic charge. Therefore, we have one electronic charge for each electron-hole pair separated in this fashion. The trapping process b-d will also contribute, for practical purposes, the same electronic charge transport if the trapped holes are near the silicon interface. If the electronic mechanism described by this model with mobile holes is to explain the relatively small quantum efficiency for the photocurrent, there must be active some other mechanism that reduce the

number of electron-hole pairs separated per incident photon. One such mechanism is geminate recombination. This is the inverse of the process a-b, which occurs when the electron becomes thermalized while still within the sphere of influence of the hole. This mechanism has been suggested as an explanation for the current-field dependence in electronic bombardment experiments [4] in SiO_2 and in vacuum ultraviolet radiation studies [7] of SiO_2 . The coulomb capture radius (Onsager radius) for Al_2O_3 is approximately 40 \AA , the phonon scattering length [10] is about 4 to 5 \AA , and the optical phonon energy [11] is about 0.1 eV. Hence, a 1-eV electron would have to suffer approximately ten phonon collisions to thermalize. Therefore, assuming isotropic scattering, it is highly probable that even in the presence of a field such an electron might thermalize within 40 \AA of the hole and thus be recaptured. Another possible explanation for the reduced electron-hole pair separation in Al_2O_3 is the mechanism of either defect or band-to-band recombination following separation. In this process, an electron separates from its parent hole but recombines with another further downstream. Since Al_2O_3 is known to have a large number of both hole and electron traps which might act as recombination centers, this process cannot be discounted.

The reduced quantum efficiency for photocurrents in aluminum oxide can be explained with another model. In this model, holes are assumed to be immobile, and the various electronic processes involved are illustrated in Fig. 9(b). The photocurrent contribution results from process a-b, the electron-hole pair generation followed by process a-c, the sweepout of electrons. The reduced apparent quantum efficiency for the photocurrent results from the fact that each electron-hole pair produces only a small fraction of an electronic charge in the external circuit, that fraction being the distance a-c divided by the thickness of the oxide. Continuity is maintained after some space charge accumulation by the injection of electrons from the silicon, some of which annihilates trapped holes by the process h-j-g. Upon cessation of the radiation, the only component that disappears immediately is the component a-c. The remaining processes such as h-j-g and h-j-k contribute to the dark current remaining when the light is turned off. Note the distinction between the

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two models in Fig. 9. In both models, the dark current is contributed by an electron injection mechanism h-j-k, but in the model of Fig. 9(a) the smaller photocurrents are explained by a recombination mechanism following the electron-hole pair generation; in Fig. 9(b), the smaller photocurrent is explained by the distance traveled by electrons in escaping from the insulator film. It should be noted that the model of Fig. 9(b), because of the shallow absorption of the vacuum UV radiation, will always yield a quantum efficiency significantly less than 100%, when the oxide thickness is much greater than the absorption depth of the radiation. On the other hand, the model of Fig. 9(a) can explain either a reduced quantum efficiency or a quantum efficiency approaching 100%, depending upon the relative magnitudes of the generation process a-b and its inverse due to recombination. With the present experimental data on aluminum oxide, it is impossible to distinguish between the two models since either can explain the reduced quantum efficiency observed. It is important to note, however, that in the case of SiO_2 where the quantum efficiency approached 100%, only the model of Fig. 9(a) can explain the experimental results.

IV. CONCLUSIONS

Radiation-induced currents in aluminum oxide using strongly absorbed vacuum UV radiation have been measured. These currents show behavior similar to that which has been observed previously in SiO_2 films; namely, there is a photocurrent component and a dark current component that is apparently produced by the tunneling of electrons into the film from the silicon electrode. Several differences are noted between these results and those reported for SiO_2 . First, the photocurrent component is much smaller than in the case of SiO_2 , resulting in significantly less than 100% quantum efficiency. Secondly, the current enhancement mechanism occurs with a much lower applied field than in the case of SiO_2 . The mechanism for the current enhancement in Al_2O_3 is believed to be the trapping of positive charge (holes) in the aluminum oxide film. Two possible models have been presented, one in which holes are mobile and one in which they are immobile. Either model can explain the presently observed experimental results. In either case, under irradiation the accumulation of positive charge in Al_2O_3 continues until the interface field is sufficient to produce tunnel injection from the silicon. This occurs at a much lower field in Al_2O_3 than in SiO_2 owing to the trap-assisted tunneling mechanism that has been described previously. Analyses of the photocurrents in this experiment have shown that it is not possible to distinguish between a model in which holes are immobile and the photocurrent results from merely the sweepout of electrons [as described in Fig. 9(b)] and a model in which holes are mobile and traverse the oxide [as in Fig. 9(a)]. Interpretation of the results in Al_2O_3 are complicated by the presence of large electron trapping and the relatively low injection threshold for electrons. The presence of both electron and hole trapping in Al_2O_3 and the difficulty in determining the location of the space charge by etching techniques make it very hard to distinguish between models involving mobile holes and immobile holes. Therefore, until some new experimental techniques are developed, it appears impossible to distinguish between the two models.

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CHIEF
LIVERMORE DIVISION FLD COMMAND DNA
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P.O. BOX 808
LIVERMORE, CA 94550

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REDONDO BEACH, CA 90278

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SAN BERNARDINO, CA 92402

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SAN BERNARDINO OPERATIONS
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SAN BERNARDINO, CA 92402

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ONE SPACE PARK
REDONDO BEACH, CA 90278

TRW SYSTEMS GROUP
ATTN EARL W ALLEN
SAN BERNARDINO OPERATIONS
P.O. BOX 1310
SAN BERNARDINO, CA 92402

TRW SYSTEMS GROUP
ATTN JERRY I LOHELL
ONE SPACE PARK
REDONDO BEACH, CA 90278

DIRECTOR
NAVAL RESEARCH LABORATORY
ATTN CODE 6631 JAMES C RITTER
WASHINGTON, DC 20375

COMMANDER
NAVAL SEA SYSTEMS COMMAND
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NAVY DEPARTMENT
WASHINGTON DC 20362

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NAVAL RESEARCH LABORATORY
ATTN CODE 4004 EMANUEL L BRANCATO
WASHINGTON, DC 20375

COMMANDER
NAVAL SEA SYSTEMS COMMAND
ATTN SEA-9931 RILEY B LANE
NAVY DEPARTMENT
WASHINGTON DC 20362

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NAVAL RESEARCH LABORATORY
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WASHINGTON, DC 20375

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NAVAL SURFACE WEAPONS CENTER
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WHITE OAK, SILVER SPRING, MD 20910

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NAVAL RESEARCH LABORATORY
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COMMANDER
NAVAL SURFACE WEAPONS CENTER
ATTN CODE 431 EDWIN R DEAN
WHITE OAK, SILVER SPRING, MD 20910

DIRECTOR
NAVAL RESEARCH LABORATORY
ATTN CODE 5210 JOHN E DAVEY
WASHINGTON, DC 20375

COMMANDER
NAVAL SURFACE WEAPONS CENTER
ATTN CODE 4450 JOHN H MALLEY
WHITE OAK, SILVER SPRING, MD 20910

DIRECTOR
NAVAL RESEARCH LABORATORY
ATTN CODE 2627 DORIS R FOLEN
WASHINGTON, DC 20375

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NAVAL SURFACE WEAPONS CENTER
ATTN CODE 4421 TECH LTR
WHITE OAK, SILVER SPRING, MD 20910

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SEATTLE, WA 98124

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PASADENA, CA 91103

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SOUTHFIELD, MI 48076

CALIFORNIA INSTITUTE OF TECHNOLOGY
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JET PROPULSION LABORATORY
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PASADENA, CA 91103

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RESEARCH LABORATORIES DIVISION
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SOUTHFIELD, MI 48076

FLORIDA UNIVERSITY OF
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GAINESVILLE FL 32611

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ORLANDO FL 32302

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PHILADELPHIA PA 19101

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SPACE DIVISION
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GODDARD BLVD KING OF PRUSSIA
P.O. BOX 8555

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SPACE DIVISION
VALLEY FORGE SPACE CENTER
GODDARD BLVD KING OF PRUSSIA
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PHILADELPHIA PA 19101

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RE-ENTRY & ENVIRONMENTAL SYSTEMS DIV
P.O. BOX 7722
3198 CHESTNUT STREET
PHILADELPHIA PA 19101

GENERAL ELECTRIC COMPANY
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P.O. BOX 7722
3198 CHESTNUT STREET
PHILADELPHIA PA 19101

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816 STATE STREET (P.O. DRAWER QQ)
SANTA BARBARA CA 93102

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SYRACUSE, NY 13201

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WESTGATE RESEARCH PARK
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MCLEAN, VA 22101

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FRENCH ROAD
UTICA, NY 13503

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ELECTRONICS SYSTEMS GRP-EASTERN DIV
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NEEDHAM, MA 02194

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FRENCH ROAD
UTICA, NY 13503

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ELECTRONICS SYSTEMS GRP-EASTERN DIV
77 A STREET
NEEDHAM, MA 02194

GENERAL ELECTRIC COMPANY
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P.O. BOX 5000
BINGHAMTON, NY 13902

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ATTN JAMES A WALTON
ELECTRONICS SYSTEMS GRP-EASTERN DIV
77 A STREET
NEEDHAM, MA 02194

GENERAL ELECTRIC COMPANY-TEMP
ATTN WILLIAM ALFONTE

GTE SYLVANIA, INC.
ATTN PAUL R FREDRICKSON
189 B STREET
NEEDHAM HEIGHTS, MA 02194

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C/O DEFENSE NUCLEAR AGENCY
WASHINGTON, DC 20305

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SANTA BARBARA, CA 93105

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NEEDHAM HEIGHTS, MA 02194

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189 R STREET
NEEDHAM HEIGHTS, MA 02194

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189 R STREET
NEEDHAM HEIGHTS, MA 02194

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MELBOURNE, FL 32901

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MELBOURNE, FL 32901

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MINNEAPOLIS, MN 55413

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GOVERNMENT AND AERONAUTICAL
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MINNEAPOLIS, MN 55413

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13350 U.S. HIGHWAY 19
ST. PETERSBURG, FL 33734

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ST. PETERSBURG, FL 33734

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COLORADO SPRINGS, CO 80933

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LOS ANGELES, CA 90009

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ORLANDO, FL 32805

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MARTIN MARIETTA AEROSPACE
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LOCKHEED MISSILES & SPACE CO INC
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DENVER, CO 80201

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PALO ALTO, CA 94304

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DENVER DIVISION
P.O. BOX 179
DENVER, CO 80201

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DALLAS, TX 75222

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DENVER DIVISION
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DENVER, CO 80201

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DALLAS, TX 75222

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SANTA BARBARA, CA 93101

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7650 CONVOY COURT
SAN DIEGO, CA 92111

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1 RESEARCH PARK
PALOS VERDES PENINSULA, CA 90274

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P.O. BOX 208
BEDFORD, MA 01730

NORTHROP CORPORATION
ATTN ORLIE L CURTIS JR
NORTHROP RESEARCH AND TECHNOLOGY CTR
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HAWTHORNE, CA 90250

R & D ASSOCIATES
ATTN S CLAY ROGERS
P.O. BOX 9695
MARINA DEL REY CA 90291

NORTHROP CORPORATION
ATTN DAVID W POCOCK
NORTHROP RESEARCH AND TECHNOLOGY CTR
3401 WEST BROADWAY
HAWTHORNE, CA 90250

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NEW YORK, NY 10014

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SAN LEANDRO, CA 94577

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ATTN K H ZAININGER
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W. WINDSOR TWP
201 WASHINGTON ROAD, P.O. BOX 432
PRINCETON, NJ 08540

PHYSICS INTERNATIONAL COMPANY
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2700 MERCED STREET
SAN LEANDRO, CA 94577

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FRONT 3 CURBER STREETS
CAMDEN, NJ 08012

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542 INDUSTRIAL WAY WEST
P.O. BOX 626
EATONTOWN, NJ 07724

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ATTN RONALD J GUTMANN
P.O. BOX 965
TROY, NY 12181

ROCKWELL INTERNATIONAL CORPORATION
ATTN DENNIS SUTHERLAND
ELECTRONICS OPERATIONS
COLLINS RADIO GROUP
5225 C AVENUE NE
CEDAR RAPIDS, IA 52406

RESEARCH TRIANGLE INSTITUTE
ATTN ENG DIR MAYRANT SIMONS JR
P.O. BOX 12194
RESEARCH TRIANGLE PARK, NC 27709

ROCKWELL INTERNATIONAL CORPORATION
ATTN MILDRED A BLAIR
ELECTRONICS OPERATIONS
COLLINS RADIO GROUP
5225 C AVENUE NE
CEDAR RAPIDS, IA 52406

ROCKWELL INTERNATIONAL CORPORATION
ATTN NEURIE C MESSENGER F 61
3370 MIRALOMA AVENUE
ANAHEIM, CA 92803

SANDERS ASSOCIATES, INC.
ATTN MEL L AITEL NCA 1-3234
95 CANAL STREET
NASHUA, NH 03060

ROCKWELL INTERNATIONAL CORPORATION
ATTN JAMES E HELL H410
3370 MIRALOMA AVENUE
ANAHEIM, CA 92803

SCIENCE APPLICATIONS, INC.
ATTN LARRY SCOTT
P.O. BOX 2351
LA JOLLA, CA 92038

ROCKWELL INTERNATIONAL CORPORATION
ATTN K F HULL
3370 MIRALOMA AVENUE
ANAHEIM, CA 92803

SCIENCE APPLICATIONS, INC.
ATTN J ROBERT BEYSTER
P.O. BOX 2351
LA JOLLA, CA 92038

ROCKWELL INTERNATIONAL CORPORATION
ATTN DONALD J STEVENS FA70
3370 MIRALOMA AVENUE
ANAHEIM, CA 92803

SCIENCE APPLICATIONS, INC.
ATTN NOEL R BYRN
HUNTSVILLE DIVISION
2109 W. CLINTON AVENUE
SUITE 700
HUNTSVILLE, AL 35805

ROCKWELL INTERNATIONAL CORPORATION
ATTN T H VATES
5701 WEST IMPERIAL HIGHWAY
LOS ANGELES, CA 90009

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333 RAVENSWOOD AVENUE
MENLO PARK, CA 94025

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1090 SAN MIGUEL ROAD
CONCORD, CA 94518

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MARCUS AVENUE
GREAT NECK, NY 11020

SYSTRON-DUNNEN CORPORATION
ATTN GORDON R DEAN
1090 SAN MIGUEL ROAD
CONCORD, CA 94518

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ATTN PAUL MARAFFINO
SPERRY DIVISION
SPERRY GYROSCOPE DIVISION
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MARCUS AVENUE
GREAT NECK, NY 11020

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P.O. BOX 5474
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