Charge-Carrier Transport Phenomena in Amorphous SiO_2: Direct Measurement of the Drift Mobility and Lifetime*

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(Received 13 April 1973)

A report of the first direct measurement of the drift velocity of excess electrons is given as a function of applied field in amorphous SiO_2 using a transit-time technique. The drift mobility is found to be 20 ± 3 cm^2/V sec at 300 K, the highest known for a solid amorphous material, and it decreases with increasing temperature. These results can be explained by a model involving LO phonon scattering at high temperatures and boundary scattering at lower temperatures.

Charge transport in quartz has been of interest because of its well-known dielectric and piezoelectric properties. Recently, metal-oxide-semiconductor devices employing amorphous SiO_2 as the insulator have been the subject of intense study;[1] in addition, quartz oscillators and piezoelectric gauges both display phenomena associated with the charge transport due to dielectric breakdown or ionizing radiation.[2] Many attempts have been made to measure the transport properties.[3-6] Photocurrents due to photoemission from metals and semiconductors and direct excitation across the quartz band gap have been observed,[1] but never with the time resolution needed to separate the lifetime, recombination kinetics, and mobility of the carriers as separate parameters. This lack of experimental resolution has lead to several erroneous conclusions about the temperature and field dependence of all three transport parameters.[7] By exciting electron-hole pairs with a very short pulse (3 nsec half width) of high-energy x rays, we have been able to measure the electron lifetime and at high electric fields the transit time of electrons across 200-μm-thick samples.

The fused quartz employed in the experiments was Suprasil II (Amersil, Inc.) which has a very low metal-ion concentration. Evaporated Al electrodes were used to sandwich the fused-quartz disks which were inserted in a temperature-controlled vacuum chamber. X rays were generated from the 600-keV electron pulse from a Febetron 706 and passed through the walls of a screen room. The high-time-resolution data were obtained using a 1-GHz Tektronix 6045 FET probe and 7904 oscilloscope. More complete experimental details are published elsewhere.[8] There was no difficulty in resolving the carrier lifetime, which varied from 10 to 14 nsec depending on the sample, by monitoring the photocurrent after the x-ray pulse at low-enough fields so that a vanishingly small number of carriers were collected at the blocking contacts in the carrier lifetime. At higher fields many carriers could be made to cross the entire disk, and the photocurrent decay could be fitted to a simple formula involving the lifetime and the collection at the Al-quartz interface,

$$i = n_0 e \mu E[1 - (\mu E/d)t]e^{-t/\tau} \text{ (amps/cm}^2\text{)}, \quad (1)$$

where n_0 is the initial (uniform) concentration of carriers, which was found to be proportional to the x-ray dose, e is the electronic charge, μ is the carrier mobility in cm^2/V sec, E is the applied field in V/cm, d the crystal thickness, and τ the carrier lifetime. The drift velocity μE was found to be linear in electric field between velocities of 2×10^5 cm/sec and 1.7×10^6 cm/sec, giving a drift mobility at 298 K of 20 ± 3 cm^2/V sec. At higher fields the transit time was obscured by the x-ray pulse width, and at low fields was unobservable because of the short electron lifetime.

Onnasch,[7] in his studies of the dc conductivity of fused quartz under Co^{60} irradiation, claimed that the drift velocity saturated at 6×10^5 cm/sec, the speed of sound in fused quartz. This acoustoelectric effect has been shown to occur in some semiconductors,[9] but clearly does not occur in fused quartz as the direct measurements indicate. The drift-velocity curve is consistent with the predictions made by Thornber and Feynman[10] and Lynch[4,6] that the drift velocity will not saturate until about 10^7 cm/sec on the basis of LO phonon scattering.

The carrier lifetime was found to be independent of temperature in the range 110 to 370 K and independent of field up to about 4×10^4 V/cm, where the electron transit time would obscure the observation of small changes in lifetime.

Thus Onnasch's[7] model for the carrier transport, in which the lifetime was predicted to have a substantial increase with both increasing electric field and temperature, is shown to be untenable. No charge transport due to holes could be detected, and the trapped holes caused significant shortening of the electron lifetime due to recombination[11] when the accumulated dose to the sample exceeded about 20 rad. The carrier lifetime was checked after each high-field shot at a low field, and the lowest practical dose per shot was used to avoid any distortion of the data; the samples could be annealed by uv photoemission of electrons from the Al electrode into the bulk.[1]

Perhaps the most interesting result in terms of understanding the transport process is the observed temperature dependence of the mobility shown in Fig. 1. In many materials the intrinsic drift mobility is masked by trap modulation of the mobility (i.e., trapping and re-emission of the carrier) or impurity scattering, which always give drift mobilities which are lower than the intrinsic value. Trap modulation displays a sharply increasing mobility with increasing temperature because of the increased probability of emission from a trap at higher temperatures. However, the data in Fig. 1 show a very clear decrease in the mobility with increasing temperature, which is suggestive of intrinsic processes involving phonon scattering.

There have been a number of treatments of the effect of LO phonon scattering in polar crystals on the electron mobility.[10,12-14] The different approaches have been summarized and applied to amorphous SiO_2 by Lynch,[6] who found that in spite of a wide variety of theoretical approaches to the problem, the predictions of the mobility, for drift velocities giving electron energies below the LO-mode energy, came out much the same. The mobility reflects the energy of the phonon mode $\hbar\omega_l$, the electron-lattice coupling constant α, and the electron effective mass m_e:

$$\mu = \frac{tr}{2m_e}\frac{1}{\alpha\hbar\omega_l}\frac{e}{2\hbar\omega_l}\frac{3kT}{}\exp\left(\frac{\hbar\omega_l}{kT}\right). \qquad (2)$$

Lynch has calculated μ for each of the measured LO modes in quartz, and found an overall mobility of 32 cm^2/V sec from Eq. (2) at 300 K, assuming an effective electron mass equal to the free mass. The theoretical curve in Fig. 1 is Eq. (2) with an effective mass of 1.4 and it can be seen that the agreement is satisfactory for 200 to 400 K; below 200 K another process appears to be limiting the mobility to about 40 cm^2/V sec.

Another vital aspect of the charge-carrier dynamics in insulators is the processes which occur in the generation of the electron-hole pair. The importance of solid-state radiation detectors and analyzers has led to much work in defining the way in which the energy deposited by the ionizing radiation is distributed in the lattice. Extensive experimental data on intrinsic semiconductors (very high mobility) has borne out the theoretical prediction[15] that the energy required to create an electron-hole pair is roughly W(eV) $\cong 1$ eV + 3 × (band gap), which for the large band gap in SiO_2[1] (~9 eV) would be about 28 eV per electron-hole pair. Because of the blocking contacts the carrier yield is reflected in the total charge observed in the external circuit [i.e., the integral of Eq. (1)]. From Eq. (1) the total charge collected should be[8]

$$Q = n_0 e d\frac{\bar{x}}{d}\left[1+\frac{\bar{x}}{d}\exp\left(-\frac{d}{\bar{x}}\right)-\frac{\bar{x}}{d}\right](\mathrm{C/cm^2}), \qquad (3)$$

where $\bar{x}$ is defined as the product $\mu\tau E$, sometimes called the range of the carrier. Because μ and τ have been determined independently from the experiment, the charge measurement as a function of E, given in Fig. 2, reveals the magnitude of n_0, the concentration of carriers/rad (28 eV/carrier translates to 4.5×10^{12} carriers/cm^3 rad). Attempts to fit the data with Eq. (3) revealed that at low fields, W (assuming that the holes contribute no current) was about a factor of 3.6 greater than the predicted 28 eV, and that the generation efficiency was increasing with in-

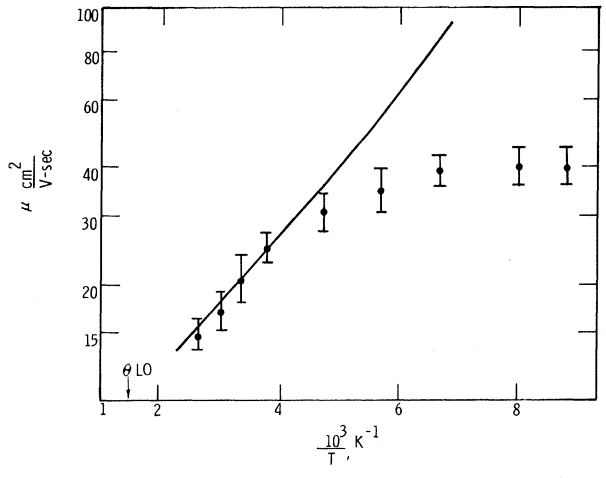

FIG. 1. Temperature dependence of the electron drift mobility. Solid line, from the Thornber-Feynman theory of electron scattering from LO phonons. The arrow indicates the Debye temperature θ_{LO}.

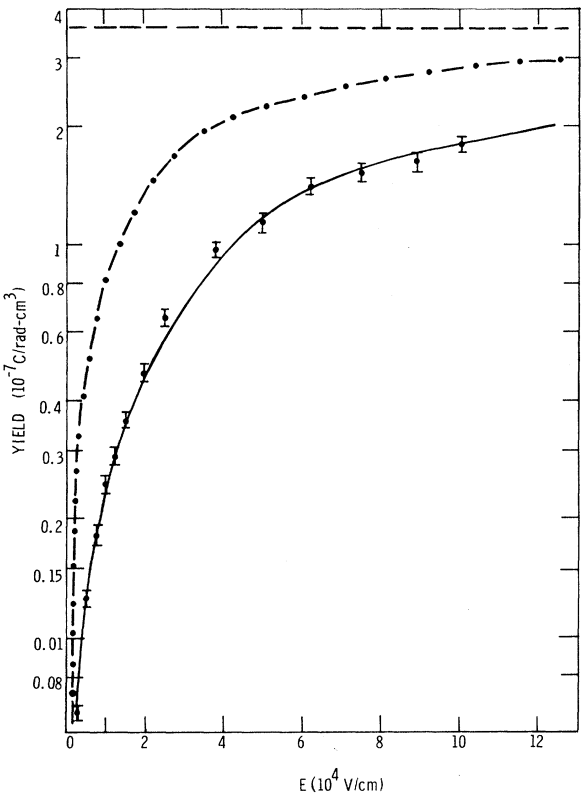

FIG. 2. The charge collected as a function of applied field. Dashed line at the top, the charge which would be collected for long-lifetime electrons assuming 28 eV per electron-hole pair; dot-dashed line, expected values for the experimentally measured mobility and lifetime. Solid line through the experimental points, same as dot-dashed curve but corrected for the field dependence of the primary yield due to geminate recombination (asymptotically approaches 28 eV per pair at high fields).

creasing field. A very likely source of this effect is the geminate recombination of the electron-hole pairs, which has been shown to be an important process in the low ionization yield of high-pressure gases,[16] and organic liquids and solids.[8,17,18] It is thought that geminate recombination will not occur in high-mobility, high-dielectric-constant semiconductors because the mean free path of the carriers is very much greater than the Coulomb capture radius (Onsager radius).[19] The carrier mobility (and presumably the mean free path) we have measured in quartz is higher than in solids, where the existence of geminate recombination has been demonstrated, but since the Coulomb capture radius $r_C = 2e^2/3\epsilon kT$ is about 100 Å ($\epsilon = 3.8$), it is entirely reasonable that a large number of geminate pairs will undergo recombination. The solid line in Fig. 2 is a plot of Eq. (3) corrected for geminate recombination,[20] assuming an initial electron-hole pair separation of 100 Å, which is consistent with our observed temperature dependence of the ionization yield. In any case, the field dependence of the yield is small compared to that observed in some of the organic systems,[8] where the initial separation is found to be much smaller.

In summary, we have found that the carrier mobility in a very pure fused quartz appears to be dominated only by the strong interactions of the excess electron with the LO phonons, giving the highest mobility measured thus far for an amorphous solid. The amorphous nature of fused quartz has been the subject of much study, and recently radial distribution functions for both x-ray and neutron scattering have indicated that crystalline order may extend out to 20 Å.[21] This is consistent with our observation that the mobility approaches a value of about 40 cm^2/V sec as the temperature is lowered; a free electron with a mean free path of about 40 Å would exhibit a mobility of that magnitude. It should be noted that the temperature dependence of the mobility can be fit by the random-phase model (RPM) which predicts a diffusing carrier with the standard T^{-1} temperature dependence.[22] The mobility is too high for a diffusing polaron,[22] but if the transfer integral J between sites is large enough, Mott[23] predicts that mobilities up to 100 cm^2/V sec can be accomodated in RPM. However, the same model predicts that there will be many localized states in the band gap which are not observed in our experiment (the 14-nsec lifetime should be very long compared with the time it should take to scatter into the localized states).

The author gratefully acknowledges helpful discussions with C. H. Seager and D. Emin and the technical assistance of David Evans.

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*Work supported by the U.S. Atomic Energy Commission.

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Observation of the Decay $K_L^0 \to \mu^+\mu^-$†

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(Received 16 April 1973)

An experiment performed at the Brookhaven National Laboratory alternating-gradient synchrotron has yielded six events above negligible background which satisfy criteria for the decay $K_L^0 \to \mu^+\mu^-$. The K_L^0 flux, measured by means of the decay $K_L^0 \to \pi^+\pi^-$, leads to a value for the branching ratio $\Gamma(K_L^0 \to \mu^+\mu^-)/\Gamma(K_L \to \text{all}) = 11 \times 10^{-9}$.

The decay $K_L^0 \to \mu^+\mu^-$ is expected to occur with a branching ratio of at least 6×10^{-9}.[1] This prediction is based on unitarity, the measured $K_L^0 \to \gamma\gamma$ rate, quantum electrodynamics, and the assumption that contributions from intermediate states other than $K_L^0 \to \gamma\gamma$ are negligible. Theoretical estimates of the maximum interference possible from other likely intermediate states do not reduce this lower bound substantially.[2] The experiment of Clark *et al.*,[3] however, places an upper limit of 1.8×10^{-9} (90% confidence level) on this decay, a value incompatible with this prediction and difficult to resolve theoretically without introduction of new particles or interaction mechanisms. The present work was undertaken as a check of the experimental result of Clark *et al.*

The apparatus was situated in a long-lived neutral beam derived from the G-10 internal target of the Brookhaven National Laboratory alternating-gradient synchrotron. Three collimators defined a solid angle of 18 μsr at an angle of 4.7° to the circulating proton beam. 8 radiation lengths of lead near the first collimator converted γ rays and two sweeping magnets eliminated charged particles from the beam. The final 13 m of the beam line, as well as the 6-m decay region, were evacuated.

K_L^0 decays were detected with a spectrometer (Fig. 1) employing three X-Y multiwire proportional chambers (MWPC). The chambers (5000 wires altogether) have 2 mm spacing between signal wires and the left and right halves of the horizontal wires are divided to allow independent readout. The spectrometer magnet was operated at 210.6 MeV/c transverse momentum, between the maximum values possible for $\mu\mu$ and $\pi\pi$ decays. Electrons were detected in an atmosphere-pressure hydrogen-gas Cherenkov counter with twelve independent optical sectors. Following the spectrometer are three walls of heavy concrete, total thickness 900 g/cm^2, and three