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Optical modulation of low-energy-electron reflection and transmission at a Si surface

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This paper reports the optical modulation of both the transmitted and backscattered currents which result when a primary beam of low-energy electrons is incident on an Ar-bombarded Si (111) surface. It is shown that the dependence of these optically modulated currents on electron energy can be separated into two components. The first component consists of a sharp peak at low incident-electron energies and is correlated with the photovoltaic modulation of the contact-potential difference between the electron gun and Si target. The second component is attributed to the photovoltaic modulation of the escape probability for secondary electrons which are created by the incident primary electrons. A theoretical model based on this physical interpretation correctly predicts the qualitative dependence of optically modulated currents on a number of experimental parameters such as incident-electron current, electron energy, incident light, and sample temperature.

I. INTRODUCTION

A number of experimental techniques examine the interactions which occur when an electron is incident on a solid surface. An incident electron can be elastically reflected or it can undergo a number of inelastic interactions. The inelastically scattered electrons can either be completely thermalized or can escape from the surface. The latter process is called secondary-electron emission[1] and is sensitive to the exact nature of the solid surface.[2] In fact, electron-solid interactions form the basis of a number of surface probes such as low-energy-electron diffraction (LEED) and low-energy-electron loss spectroscopy (LEELS).

Recently, it has been demonstrated that when low-energy electrons are incident on a semiconductor surface, the transmission of electrons into the bulk can be optically modulated.[3,4] In the initial experiments, the external voltage difference between the semiconductor and electron-gun cathode was chosen to coincide with the sharp increase in the total current transmitted through the semiconductor as a result of incident electrons. The work function of the semiconductor was varied by incident light due to photovoltaic changes in the band bending, and this resulted in the optical modulation of the transmitted current. This experimental approach is a variation of the standard retarding-potential method for measuring changes in the work function. These first experiments were performed at a relatively low potential difference between the gun and the semiconductor because the initial theoretical model predicted that at higher potential differences there would be little or no optical modulation of the transmitted current.[3,4] When higher potential differences were examined, however, it soon became apparent that this theoretical model was incorrect.[4-6] It has been suggested that the observed optical modulation of the current at these higher potentials is due to the photovoltaic modulation of the secondary-electron emission.[5] The analogous physical process of optical modulation of photoemission is known to occur.[7-9]

In all of the previous experiments,[3-6] only the optically modulated component of the transmitted current was measured. In this paper the existing experimental apparatus has been modified to include a collection grid so that complementary measurements can be made of the optical modulation of both the transmitted and backscattered currents. The dependence of both optically modulated currents on incident-electron current, electron energy, incident light, and sample temperature is reported. A theoretical model which involves secondary-electron emission at the higher incident-electron energies is presented which adequately explains the observed experimental results.

II. EXPERIMENTAL

The experimental apparatus has been described in detail in a previous publication,[4] and consists of a low-energy-electron gun and a semiconductor sample holder in an ultrahigh-vacuum system. Electrons are thermionically emitted from a filament of W wire which is resistively heated by a 1.5-V floating power supply. The positive terminal of this supply is floated at a variable negative potential from ground thereby defining the electron-gun voltage V_g. A metal grid has been placed between the electron gun and the semiconductor sample. In conventional studies of backscattered electrons, care has to be taken to shield collector grids from incident primary electrons. This precaution was unnecessary in the present experiments since the primary electron beam does not have an optically modulated component, while only the optically modulated component of the grid current is detected by the lock-in amplifier. Both the metal grid and the semiconductor sample are grounded through one of several low-impedance detector circuits. The total current transmitted through the semiconductor is measured by a microammeter. The optically modulated component of this current, resulting from the chopped light, is measured either by phase-sensitive lock-in detection or by a signal averaging detector. The ratio of the optically modulated component of the transmitted current to the total transmitted current is of the order of 1 part in 10^6. Since this is negligibly small, the total transmitted current is approximately the same with or without the chopped incident light. In the present work, however, the standard procedure of measuring the total transmitted current in the dark is employed.

Both p (0.4–0.6 $\text{ohm}^{-1}\,\text{cm}^{-1}$) and n (2 $\text{ohm}^{-1}\,\text{cm}^{-1}$) type Si(111) samples were used in this work. Both faces of the Si samples were Syton polished and an indium contact was made to the rear face prior to placement in the ultrahigh-vacuum system. The samples were approximately 1 mm thick, which is large compared to the penetration depth of the above band-gap light employed. Therefore, the possible interference from optical modulation of the properties of the rear In contact can be ignored. After bakeout, the base pressure was less than 5×10^{-10} Torr. The samples were subjected to an Ar-bombardment cleaning prior to all experimental measurements (3 keV, 30 mA, $\frac{1}{2}$ h, 6×10^{-5} Torr Ar). Ar bombardment did not appreciably change the magnitude of the optically modulated currents for the p-type Si. This is in contrast to the results for n-type Si where Ar bombardment reduced the optically modulated current below the detection limit. The reason for this difference between the n- and p-type Si is not understood at present. The figures in this paper are based on the experimental results for p-type Si.

White light from a xenon lamp was employed, providing a photon flux of approximately 10^{13} photons $\text{cm}^{-2}\,\text{sec}^{-1}$ at the sample. At higher light intensities, it was determined that the photoemission component became an appreciable fraction of the optically modulated currents. Photoemission could be separated from effects due to the primary electron beam since the former is present when there is no incident primary electron beam. For the work reported here, the component due to photoemission was less than 0.1% of the optically modulated current and therefore was ignored. The incident light was chopped at 28 Hz, and it was determined that the response of the current to the changes in incident light was fast compared to this chopping period.

When the retarding-potential electron-beam technique is used to determine work-function values, the experimental geometry is usually designed so that the voltage drop across the emitting filament can be neglected.[10,11] The transmitted current as a function of the external voltage on the emitting cathode goes through a sharp rise before reaching a constant saturation current. For this geometry, the sharpness of the rise in the current is determined by the thermal spread of the emitted electrons, and the external voltage where the break in the transmitted current occurs, commonly referred to as the "knee," is given by[10]

$$V_{\text{ext}}=\phi_s-\phi_c\,, \tag{1}$$

where ϕ_s is the work function of the sample and ϕ_c is the work function of the emitting cathode. Equation (1) implies that accurate measurements of V_{ext}, the external voltage at the "knee," can be used to determine the contact potential difference between the sample and emitting cathode.

The experiments reported in this paper are similar except that the geometry of the electron gun is such that electrons from the entire length of the filament are incident on the sample. Consequently, the voltage drop across the filament cannot be neglected. The emitted electrons from the electron gun therefore have an energy spread which is determined both by the thermal spread of the emitted electrons and by the voltage drop across the electron-gun filament. This energy spread can be determined directly using the standard "stopping potential" analysis.[12] The current which was transmitted through a gold foil was measured as a function of the electron-gun voltage; the results were numerically differentiated and are shown in Fig. 1. It should be noted that, for this one case, the gold foil was negatively biased in order to offset the curves along the V_g axis for reasons of convenience. The results of Fig. 1 indicate that the emitted electrons have an energy spread full width at half maximum of 3.3 eV. When the additional negative biasing on the gold foil was removed, the "knee" of the transmitted current curve shifted to a gun voltage of 3.6 V. From the literature values of ϕ_{Au} and ϕ_{W} (see Ref. 13) and Eq. (1), it would be expected that the "knee" would occur at 0.7 V. The shift of the position of the "knee" by 2.9 V is consistent with the measured energy spread of the emitted electrons.

Because of the voltage drop across the gun filament, the voltage of the transmitted current "knee" is no longer equal to the contact potential differ-

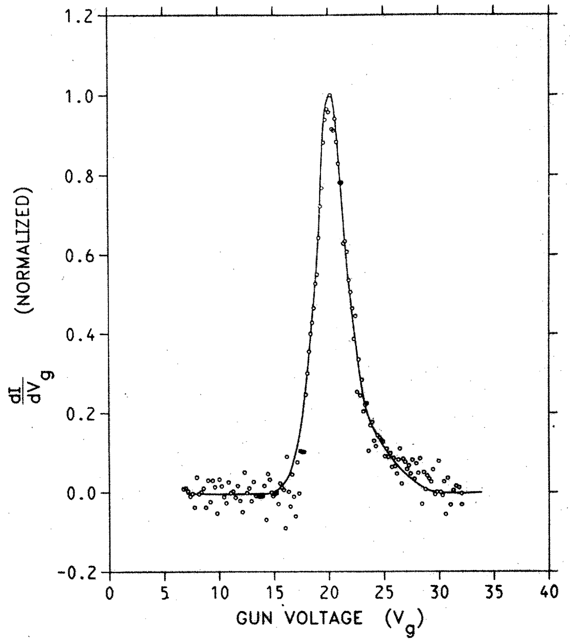

FIG. 1. Stopping-potential curve for electron gun and gold sample which gives the energy distribution of the emitted electrons (see text).

ence. Since the voltage drop across the filament is a constant, however, shifts in the position of the knee can be used to measure relative changes in the work function. For example, when the Au sample was replaced with the Si sample which is used for all the remaining figures in this paper, the "knee" of the curve shifted to 2.8 V. Using the literature value for ϕ_{Au} (5.32 eV),[13] this implies that the work function of this sample was approximately equal to 4.5 eV. This is consistent with literature values for ϕ_{Si} which range from 4.55 to 5.15 eV.[13] A number of Si samples have been examined and they all exhibited work functions within this range, with one exception. When this one sample was removed and a fresh In contact was made, the work function also fell within the expected range. The cause of the problem was therefore attributed to a poor rear contact which resulted in sample charging and a resultant shift in the "knee" of the curve.

III. RESULTS AND DISCUSSION

Normally, experiments using the electron-beam retarding-potential technique involve chemically changing the sample surface condition and measuring the resulting change in work function.[11] In contrast, recent variations of the technique involve changing the work function with incident light and observing the resultant photovoltaic changes in the semiconductor band bending.[3-6]

The physical origin of the optically modulated component of the transmitted current can readily be understood. The current transmitted through a semiconductor in the dark has a "knee" position determined by the work function of the semiconductor in the dark. When light is incident on the semiconductor, there is a photovoltaic change in the work function and this shifts the "knee" of the resulting transmitted current curve along the gun voltage axis. The first theoretical model which was proposed[3,4] assumed that the transmitted current curve would remain unchanged in form as it was shifted along the gun voltage axis. Therefore, for small photovoltage signals, the optically modulated transmitted current would simply correspond to the first derivative of the total transmitted current. As a result, the optically modulated current would be expected to consist of a single sharp peak at a gun voltage where the sharp increase in the total transmitted current is observed. This correlation has in fact been observed.[3-5] However, in addition to this sharp peak in the optically modulated current, there is also optically modulated current at relatively high external voltages for a number of semiconductors.[4-6] This second component in the optically modulated current cannot be accounted for by the theoretical models in the literature.[3,4]

One such semiconductor where the optically modulated current consists of the two components described above is Si. Figure 2 shows the optically

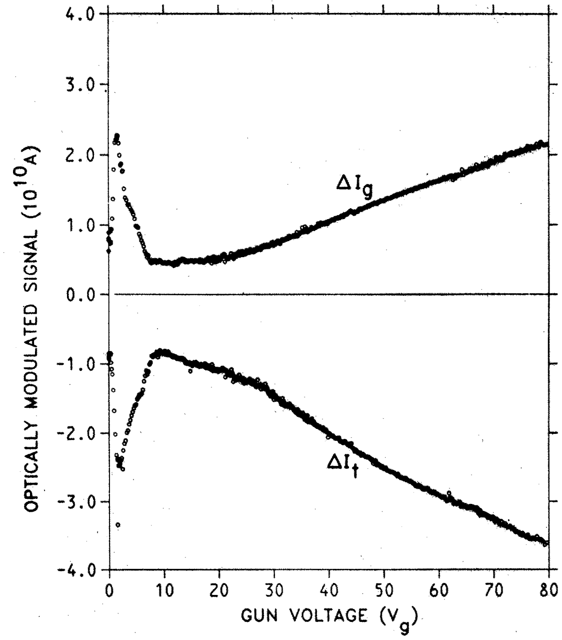

FIG. 2. Optically modulated transmitted (ΔI_t) and backscattered (ΔI_g) currents as a function of electron-gun voltage.

modulated current ΔI_t transmitted through the Si target as well as the optically modulated current ΔI_g backscattered from the Si target and collected by the metal grid. The ΔI_g signal was smaller than ΔI_t signal, and this arises from the fact that the metal grid is not a perfectly efficient collector of the backscattered electrons. These optically modulated currents are shown as a function of V_g, the electron-gun voltage. The two modulated currents have been measured by phase sensitive lock-in detection, and are 180° out of phase, as is expected on the basis of conservation of total charge. From an analysis of the time dependence of the two modulated currents using a signal averager, it was determined that incident light caused a decrease in the transmitted current and an increase in the backscattered current for the p-type Si samples. For that reason ΔI_g has been plotted along the positive y axis. The sign of this time dependence was reversed for the n-type Si. In all other respects, however, the results for n-type Si were qualitatively similar to the results for the p-type Si.

When the dependence of total transmitted current on gun voltage is numerically differentiated, the derivative consists of a single sharp peak at a low gun voltage which correlates closely with the observed peak in the optically modulated currents shown in Fig. 2. At higher gun voltages, however, the optically modulated currents in Fig. 2 are many orders of magnitude larger than would be predicted from the calculated first derivative of the total transmitted current. This discrepancy at higher gun voltages between the experimental results in Fig. 2 and the calculated derivative of the total transmitted current is an indication that one or more of the assumptions in the initial theoretical model are not valid. At the higher gun voltages it is known that the transmitted current reaches a saturation level. One of the key assumptions in the initial model was that the transmitted fraction of the current in this high gun voltage regime is independent of photovoltaic changes in the band bending.[3,4] However, this assumption oversimplifies the interaction between an electron and a solid surface because it ignores the physical process of secondary electron emission.[1,2] It is known that when an electron is incident on a solid surface it can lose energy through a variety of inelastic interactions. This inelastically scattered primary electron, or its associated secondary electrons, can be backscattered and escape from the solid surface back into the vacuum. The total amount of transmitted current is dependent on the amount of this backscattering. In the related phenomena of photoemission,[7-9] the escape probability for emitted electrons can be optically modulated by photovoltaic changes in the semiconductor work function. Therefore, a reasonable modification to the existing theoretical model would be to assume that a similar photovoltaic modulation of the secondary-electron emission can occur and is responsible for the observed optically modulated current at the higher electron-gun voltages.

The theoretical dependence of this model on incident-electron energy can readily be derived. The treatment presented here is for the one-dimensional case, but the extension to three dimensions is straightforward and preserves the significant features of the one-dimensional model.[14] Let δ represent the number of secondary electrons escaping from the surface. Then,

$$\delta = \int_0^\infty n(x, E_0) f(x)\, dx \ , \qquad (2)$$

where x is the distance into the bulk, E_0 is the energy of incident primary electrons, $n(x, E_0)$ is the density of secondary electrons created, at a depth x by the primary electrons of energy E_0, and $f(x)$ is the probability that a secondary electron at depth x can migrate to the surface and escape. Equation (2) contains the usual assumptions[14-16] that a distinction can be made between the production and escape mechanisms of secondary electrons, and that the energy distribution of the internal secondaries is unimportant. The theoretical problem then becomes one of finding suitable expressions for $n(x, E_0)$ and $f(x)$ to use in Eq. (2).

The escape probability is normally written in the form

$$f(x) = B e^{-x/L} \ . \qquad (3)$$

The term $e^{-x/L}$ expresses the probability that a secondary electron will migrate to the surface, and therefore L is interpreted as a diffusion length. B is the probability that a secondary electron will escape once it has reached the surface and would be expected to depend on the energy of the secondary electron, the work function of the surface, and the rate of electron trapping at the surface.

The theoretical expression for $n(x, E_0)$ is given by

$$n(x, E_0) = \frac{-N_0}{\epsilon}\left(\frac{dE}{dx}\right) , \qquad (4)$$

where N_0 is the number of incident primary electrons which penetrate into the surface region. This assumes that the density of secondaries is equal to the rate of energy loss with distance of the primary-electron beam dE/dx, divided by the average energy required to excite one secondary electron ϵ. Theoretical treatments differ in the expression used for dE/dx. In this paper we will use the constant-loss assumption first proposed by Young[17] which takes into account the scattering of the incident primaries. Therefore,

$$\frac{dE}{dx}=\frac{-E_0}{d} \quad . \tag{5}$$

This model assumes that the rate of energy loss of the primary electrons is constant and that therefore the number of secondary electrons created per unit path length is also constant.

In order to make use of Eq. (5), it is necessary to know the relationship between the maximum penetration depth d of the primary-electron beam and the energy E_0 of the primary-electron beam. Calculating this from first principles is difficult since a number of complicated energy-loss mechanisms are involved.[18,19] Fortunately, experimental work on secondary-electron emission provides a relatively simple empirical expression[17,20,21]

$$d=E_0^n/An \; . \tag{6}$$

This formula is referred to as the power law for primary-electron energy loss where A is a constant characteristic of the material and n is an adjustable parameter. Normally, good fits to the secondary-electron emission data are obtained in the range $1 \leq n \leq 2$.[17,20,21]

Equations (4)–(6) can be combined to give an expression for the density of secondary electrons at a depth x created by the primary-electron beam. This can then be used, along with the expression for the escape probability given in Eq. (3), to solve for the number of escaping secondary electrons for a given incident primary-electron energy

$$\delta=\int_0^d \left(\frac{N_0BAn}{\epsilon}\right)E_0^{1-n}e^{-x/L}dx \; . \tag{7}$$

Substituting Eq. (6) for the upper limit and integrating yields

$$\delta=-\left(\frac{N_0BAnL}{\epsilon}\right)E_0^{1-n}(1-e^{-E_0^n/LAn}) \tag{8}$$

On the basis of Eq. (8) it is possible to calculate the optical modulation of the secondary-electron emission. B depends on the work function of the material and therefore depends on the incident light and associated photovoltaic change in band bending. The other parameters in Eq. (8) are independent of the incident light. Therefore,

$$\Delta\delta=\delta_{\text{light}}-\delta_{\text{dark}}\propto B_{\text{light}}-B_{\text{dark}} \; . \tag{9}$$

The optical modulation of the escape probability for secondary-electron emission should be related to the analogous physical process of optical modulation of the escape probability for photoemission.[7-9] For the case of secondary-electron emission, the escape probability should be primarily dependent on the recombination of the secondary electron with holes at the top of the valence band, and/or at surface states. Therefore, the analogous process would be the optical modulation of photoemission caused by light with photon energy near the photoemission threshold. This problem has been examined in the literature[7-9] and in these studies; it should be noted that the second light beam used for the optical modulation is always below the photon-energy threshold for photoemission. For photon energies well above this threshold, it is known that a photovoltaic reduction in the work function results in an increase in the photoemitted current, as is expected from simple band-bending arguments.[7-9] At photon energies near the threshold, however, there is an unexpected reversal in sign[7,9] and in some cases,[9] photovoltaic reduction in the work function results in a *decrease* in the photoemitted current. The mechanism of the optical modulation in this latter case is not understood. Lifetime broadening,[7] and a change in the ionization energy at the surface[9] have both been suggested. Based on these results for the optical modulation of photoemission near the threshold, it will be assumed that the optical modulation of the escape probability for secondary-electron emission can be written

$$B_{\text{light}}-B_{\text{dark}}=-K\Delta\phi \; . \tag{10}$$

The photovoltaic change $\Delta\phi$ in the work function is included in Eq. (10) since it is assumed that the photomodulation of the emission is arising from photovoltaic changes in the energy band bending.

Using the above expression, the photomodulation of the secondary-electron emission is

$$\Delta\delta=+(N_0KAnL/\epsilon)\Delta\phi E_0^{1-n}(1-e^{-E_0^n/LAn}) \; . \tag{11}$$

This then predicts the dependence of the experimentally measured optically modulated signal

$$-\Delta I_t=c\Delta I_g=q\Delta\delta \; , \tag{12}$$

where c is the collection efficiency of the grid and q is the charge on the electron. The incident energy of the primary-electron beam is directly related to the electron-gun voltage.[10] Therefore, Eq. (12) leads to the following experimental dependence on the electron-gun voltage:

$$\Delta I_t=B_1V_g^{(1-n)}(1-e^{-B_2V_g^n}) \; . \tag{13}$$

Because of experimental limitations on the maximum gun voltage possible with this electron gun, it is not known if the optically modulated current reaches a plateau at higher gun voltages, or eventually decreases with increasing electron-gun voltage. For GaP, however, it is known that the optically modulated current reaches a plateau and therefore $n = 1.0$ for that material.[5] It will be assumed that n can also be set equal to 1.0 for Si. This is equivalent to assuming that the primary-electron penetration depth is linearly dependent on electron energy for these comparatively small incident-electron energies.

The qualitative origin for the exponential dependence on incident-electron energy can best be understood by substituting Eq. (6) into Eq. (11):

$$\Delta\delta = + (AK/\epsilon)N_0\Delta\phi L(1 - e^{-d/L}) . \tag{14}$$

The exponential dependence is seen to depend on d/L, the ratio of the maximum penetration depth d of the primary electrons compared to the diffusion length L of the secondary electrons. In the limit of zero primary-electron energy, d is zero from Eq. (6) and therefore the measured signal $\Delta\delta$ in Eq. (14) is zero. As the primary-electron energy is increased beyond the point where the penetration depth is significantly larger than the diffusion length of the secondary electrons, $\Delta\delta$ will reach a saturation value from Eq. (14). Additional increases in the primary-electron energy will lead to additional secondary electrons, but these secondary electrons are too deep within the bulk to be able to migrate to the surface and contribute to the observed emission.

Equation (14) correctly predicts the broad rise in the optically modulated currents at the higher gun voltages shown in Fig. 2. It does not, however, contain a term which would fit the sharp peak at low gun voltage. This is an artifact of the derivation of Eq. (14) since it was assumed that the number of primary electrons penetrating into the surface was a constant N_0. Although this is a good assumption at gun voltages above the "knee" in the transmitted current curve, it is a very poor assumption at lower gun voltages where the number of primary electrons penetrating into the surface is known to be optically modulated. However, this low gun voltage region is already adequately handled by existing theories,[3,4] and therefore the limitation of Eq. (14) to the higher gun voltage region is not a serious problem.

Equation (14) can be used to predict the experimental dependence on a number of parameters. For example, it predicts that the optically modulated current should depend linearly on N_0, the total number of incident electrons penetrating in-

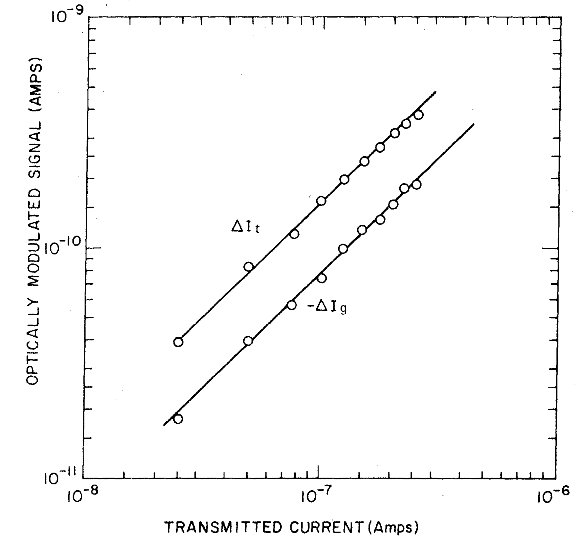

FIG. 3. Dependence of optically modulated currents on the total transmitted current at an electron-gun voltage of 50.0 V.

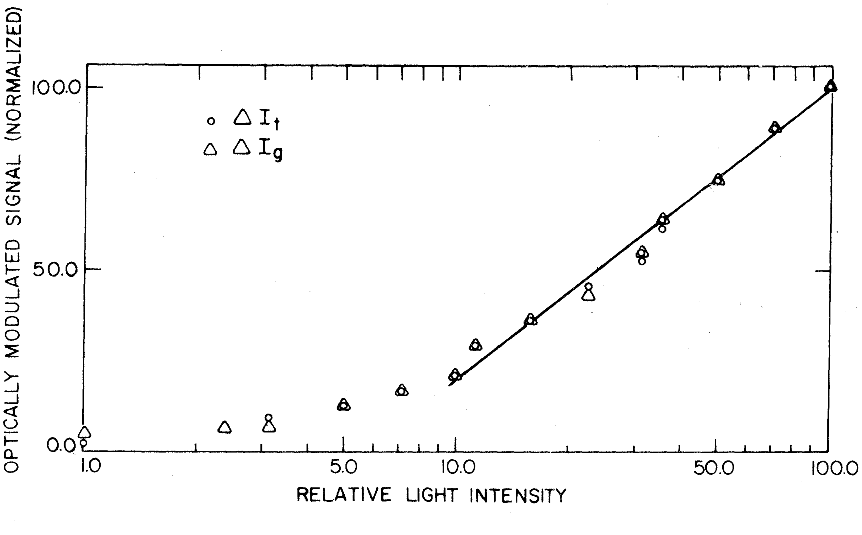

FIG. 4. Dependence of optically modulated currents on light intensity at an electron-gun voltage of 50.0 V.

to the surface region. It will be assumed that N_0 is proportional to the total transmitted current, I_t. Figure 3 shows both of the optically modulated currents, ΔI_t and ΔI_g, as a function of the total transmitted current I_t. Again, ΔI_g is less than ΔI_t because of the collection efficiency of the grid. These results were obtained at a gun voltage of 50.0 V so that Eq. (14) is applicable. Figure 3 shows the expected linear relationship predicted by Eq. (14) for an experimental range of more than two decades.

Equation (14) also predicts that the optically modulated current should depend linearly on the photovoltaic change in the work function $\Delta\phi$. This arises from the assumption made concerning the escape probability made in Eq. (10). There are two ways to experimentally check this dependence. The first makes use of the fact that $\Delta\phi$ is known to depend logarithmically on the light intensity. Again, the gun voltage has been set at 50.0 V so that Eq. (14) is applicable. The dependence of the optically modulated currents on light intensity is shown in Fig. 4 and has the expected logarithmic dependence. Deviation of the signal from the logarithmic dependence at the lower light intensities is due to the fact that the signal's time dependence is no longer a square wave at these lower light intensities. The second way to check the experimental dependence on $\Delta\phi$ in Eq. (14) is to examine the time dependence of the optically modulated currents. The sign of the photovoltaic change in the band bending can be determined by measuring the time dependence of the optically modulated transmitted current in the low gun voltage region. Incident light caused a decrease in the work function for the n-type Si, and an increase in the work function for the p-type Si. Equation (14) would therefore predict that in the high gun voltage region, incident light would cause an increase in the transmitted current for the n-type Si, and a decrease in the transmitted current for the p-type Si. This agrees with the experimental observations (Fig. 2).

At low gun voltage the size of the optically modulated current depends on the photovoltaic change in energy band bending and therefore the temperature dependence should reflect the temperature dependence of $\Delta\phi$. At the higher gun voltages on the other hand, the optically modulated current should include the temperature dependence both of $\Delta\phi$ and of the diffusion length on the basis of Eq. (14). A comparison of the two curves in Fig. 5 indicates that decreasing the temperature decreases L. Since L is proportional to the square of the steady-state photoconductivity, it is possible to compare the temperature dependence of L observed in Fig. 5 with literature values for the temperature dependence of the photoconductivity. It has been reported that for amorphous Si films,[22] the photoconductivity falls off with decreasing temperature in this temperature region. This is qualitatively consistent with the observed decrease in L computed from Fig. 5 for these Ar-bombarded Si samples.

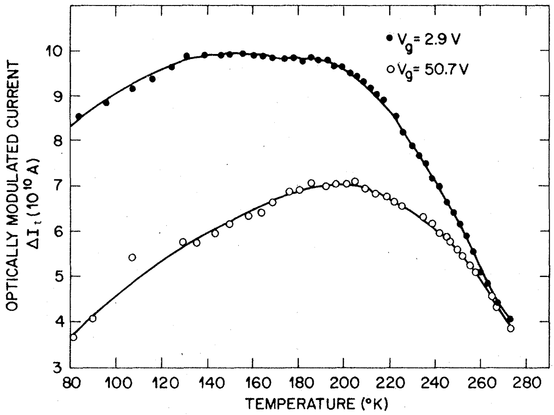

FIG. 5. Dependence of the optically modulated transmitted current on temperature at 2.9 and 50.7 V.

IV. CONCLUSION

Both the optically modulated transmitted and backscattered currents consist of the two separate components. At low gun voltages, the optically modulated changes in these currents are closely correlated with the first derivative of the total transmitted current. At higher gun voltages, optically modulated currents which are unrelated to the first derivative of the total transmitted current are observed. A one-dimensional model for this high gun voltage component involving photovoltaic modulation of the escape probability for secondary-electron emission is proposed. This model correctly predicts the qualitative dependence of the optically modulated currents on a number of experimental parameters.

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