Photoelectronic properties of high-resistivity GaAs:O †

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The photoelectronic properties of high-resistivity n-type GaAs:O have been investigated through measurements of dark conductivity and Hall effect, optical absorption, photoconductivity, and photo-Hall effect as a function of photon energy, light intensity and temperature, optical quenching of photoconductivity, and thermally stimulated conductivity. The results of all measurements can be described consistently by a four-level model (0.5, 0.7, 1.0, and 1.25 eV below the conduction band edge) plus eight electron trapping states (with depth between 0.15 and 0.54 eV) common to many different types of high-resistivity GaAs. At 82°K, illumination with photons of energy between 1.0 and 1.25 eV produces a major shift in the photoconductivity toward p type and a persistent quenching of n-type photoconductivity that recovers abruptly only upon warming above 105°K. Low-frequency photocurrent oscillations are associated with the 1.25-eV level. The capture cross section of five of the electron trapping states is small $[(5-8) \times 10^{-19} \ cm^2)]$ and decreases with electric field.

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I. INTRODUCTION

It has been known for some time that the presence of oxygen during the synthesis of GaAs produces a wide range of resistivity, from 10^{-1} to $10^8 \ \Omega \ cm$, without the material becoming p type. To explain such an automatic compensation phenomenon, it has been recognized that a deep level associated with oxygen must be involved. The position of this deep level has been sought by a number of investigations.[1-12] In general their results indicate two possible deep levels at about 0.4 and 0.75 eV below the conduction band, although a number of other levels have been reported as well.

Measurements on high-resistivity GaAs:O to date have each concentrated on a single particular analytical technique. It has been our purpose to investigate the properties of oxygen impurity in GaAs crystals by making a number of different kinds of measurement on the same samples. This work was correlated with an investigation of similar properties in high-resistivity GaAs:Cr [13] and with an investigation of electrotransmission effects in high-resistivity GaAs:O.[14,15]

High-resistivity oxygen-doped GaAs single crystals were boat grown by the Monsanto Chemical Co., Electronic Materials Division, St. Louis, Mo.[16] Attempts were made to vary the incorporated oxygen impurity concentration by varying the oxygen pressure in the reactor tube during the crystal growth with predicted variations of incorporated oxygen density between 4×10^{17} and $3 \times 10^{18} \ cm^{-3}$. Figure 1 shows the optical absorption at 300°K for five different crystals. All crystals show strong extrinsic absorption with a low-energy threshold of about 0.5 eV in the more conducting crystal, and of about 0.7 eV in the more resistive crystals. In spite of the expected variation in the concentration of incorporated oxygen in the more resistive samples, the observed absorption constant varies only slightly and in an unsystematic way with predicted oxygen density. Electrical properties of three of the samples shown in Fig. 1 are summarized in Table I.

Subsequent mass spectrographic analysis[17] of a sample, like sample 2 above, indicated an incorporated oxygen density of only $8.0 \times 10^{16} \ cm^{-3}$ with impurities

B, Al, and C also present in densities greater than $10^{16} \ cm^{-3}$ $[(1.0-7.0) \times 10^{16}, 1.2 \times 10^{16}, \ and \ 2.0 \times 10^{16},$ respectively]. It was on this sample that the photoelectronic analysis reported in this paper was carried out.

II. EXPERIMENTAL RESULTS

A. Dark conductivity and Hall effect

Between 290 and 330°K the dark conductivity has an activation energy of 0.48 eV. The Hall mobility is approximately independent of temperature in this range, but decreases strongly with decreasing temperature below 290°K in a manner inconsistent with conventional
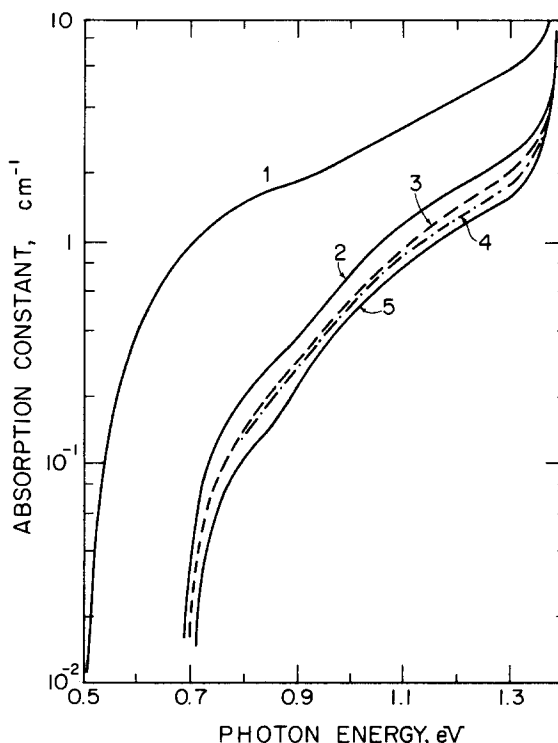

FIG. 1. Optical absorption spectra for five crystals of GaAs:O at 300°K. Numbers on the curves correspond to electrical data in Table I.

TABLE I. General properties of GaAs:O crystals.

Crystal	ρ $(\Omega$ cm)	300°K values n(cm^{-3})	μ (cm^2/V sec)	ΔE, eV (ln$n$ vs $1/T$)	Dark Fermi level (eV) $(E_c - E_F)$, 300°K
1 [a]	7.0×10^{-1}	2.0×10^{15}	4.8×10^3	0.06	0.13
5 [a]	8.0×10^5	1.5×10^9	5.4×10^3	0.43	0.49
2 [a]	3.0×10^7	2.8×10^7	7.0×10^3	0.69	0.60
Sample for detailed measurements	1.6×10^7	9.8×10^7	3.8×10^3	0.48	0.58

[a]Numbers correspond to curves of Fig. 1.

scattering processes or two-carrier effects, most likely to be associated with inhomogeneity effects in high-resistivity GaAs.[18,19] The electrical properties of this sample are also included in Table I.

B. Photoconductivity and photo-Hall effect

The spectral response curves for photoconductivity and photo-Hall mobility at 295 and 213 °K are shown in Fig. 2. Over this temperature range there is little variation in the general character of the spectral response. In addition to the photoconductivity peak for band-gap excitation at 1.4 eV, a low-energy threshold at about 0.7 eV and an indication of structure at about 1.0 eV are found in the extrinsic region. The photo-Hall mobility is n type at all photon energies, indicating that extrinsic excitation is from a level 0.7 eV below the conduction band, and possibly from a level 1.0 eV below the band as well.

The spectral response curves measured at 82 °K are distinctly different. Because very long time constants (exceeding several hours) were found over part of the spectral range, non-steady-state spectral response was measured where necessary. Figures 3(a) and 3(b) show the spectral response for photoconductivity and photo-Hall mobility as measured 3 and 15 min after onset of photoexcitation at each selected photon energy in two different ways: (a) measuring from high to low photon energy on a previously heated and cooled (in the dark) crystal, and (b) measuring from low to high photon energy on a previously heated and cooled (in the dark) crystal.

These results can be interpreted to indicate two "states" for the crystal: a higher-sensitivity n-type state, encountered if the sample has not been previously exposed to photons in the 1.0—1.3-eV range, and a lower-sensitivity p-type state to which the higher-sensitivity n-type state is reduced by exposure to photons in the 1.0—1.3-eV range. The photoconductivity in the p-type state is about two orders of magnitude smaller than in the n-type state. For measurements from low to high photon energy, 3- and 15-min data are about the same for photon energies between 0.74 and 1.0 eV, indicating that a steady state is reached by 3 min. In this region the photo-Hall mobility is n type and relatively constant, indicating that a transition from a level 0.74 eV below the conduction band to the conduction band is dominant in this region. In the high-to-low photon energy measuring sequence, a strong extrinsic photoconductivity is observed for photon energies between 1.3 and 1.5 eV, which falls rapidly for

energies less than 1.25 eV. The corresponding photo-Hall mobility in this region is n type and almost constant. These results suggest an excitation from a level at about 1.2—1.3 eV below the conduction band, which cannot be resolved at room temperature.

The temperature dependence of photoconductivity for intrinsic photoexcitation (at the intrinsic photoconductivity maximum between 1.4 and 1.5 eV) and the corresponding photo-Hall mobilities are shown in Figs. 4(a) and 4(b). Curve 1 represents the crystal in the high-sensitivity state; at low temperatures the ln(photo-conductivity) increases with $1/T$ with a slope corresponding to 0.066 eV. It is shown in the accompanying
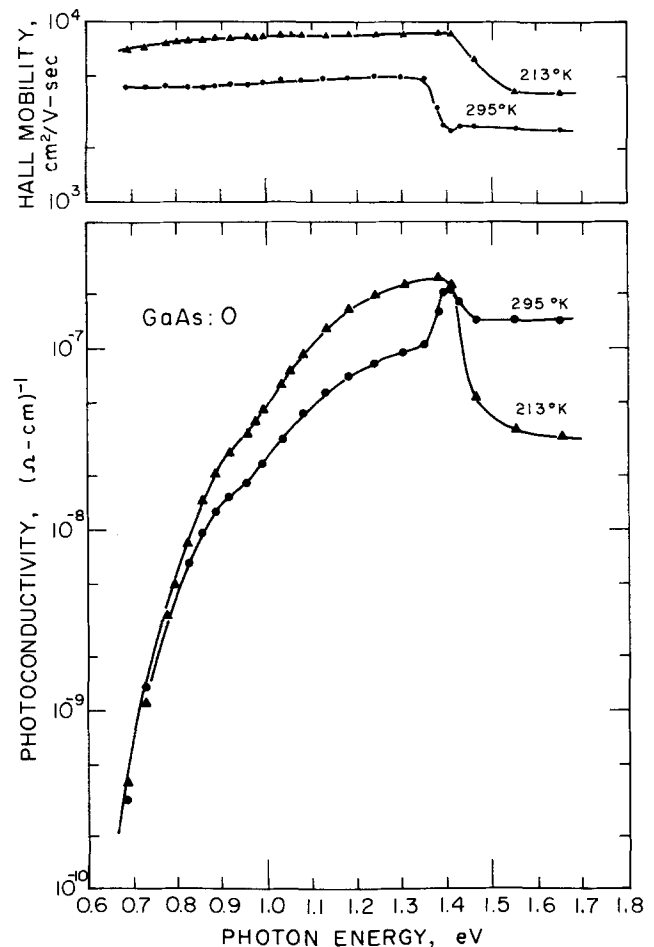

FIG. 2. Photoconductivity spectral response and variation of photo-Hall mobility with photon energy for GaAs:O at 295 and 213 °K.
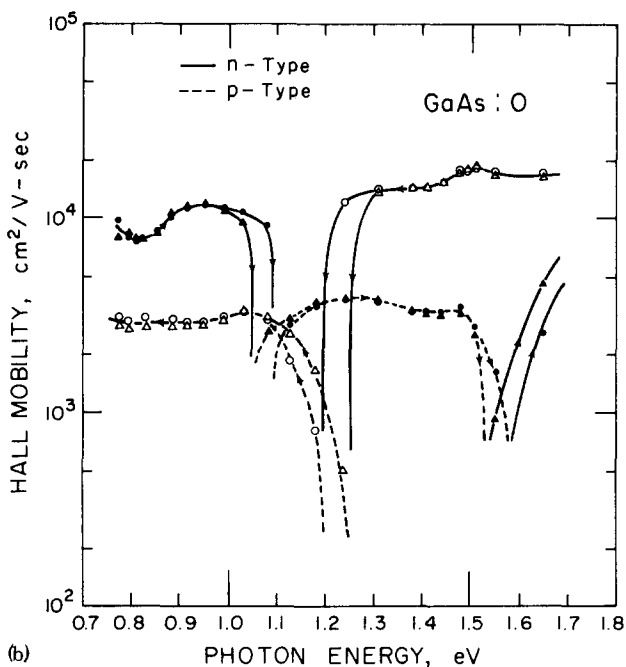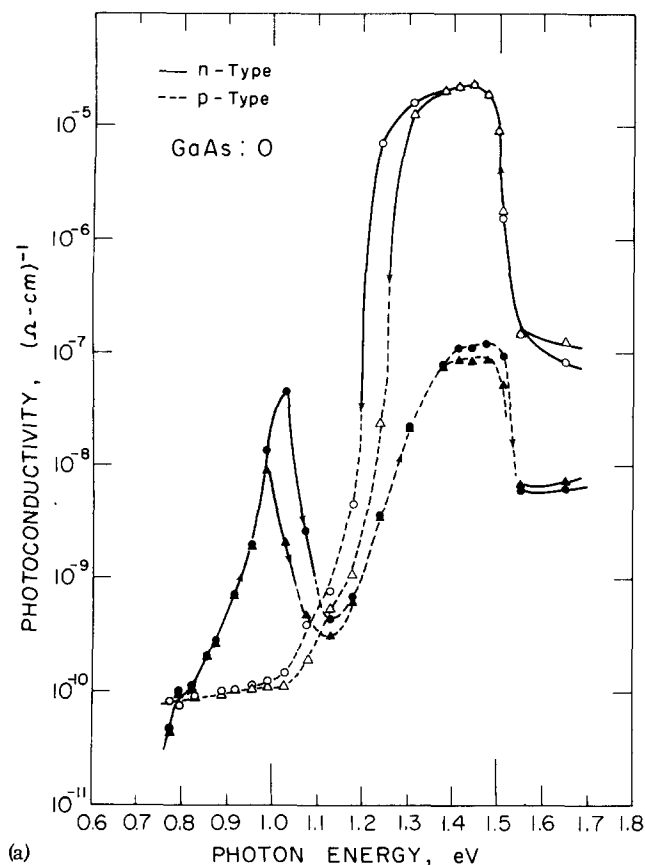
FIG. 3. (a) Photoconductivity spectral response curves measured at 82 °K for GaAs :O: measured from high to low photon energies, reading after 3 min (○) and after 15 min (△); measured from low to high photon energies, reading after 3 min (●) and after 15 min (▲). (b) Photo-Hall mobility vs photon energy for GaAs :O at 82 °K. Symbols are the same as in (a).

paper on GaAs :Cr [13] that this is the behavior expected from the Shockley-Read one-level recombination model for a level about 0.65 eV below the conduction band (0.066 eV below the Fermi level) with $\tau_{n0} = 6 \times 10^{-10}$ sec.
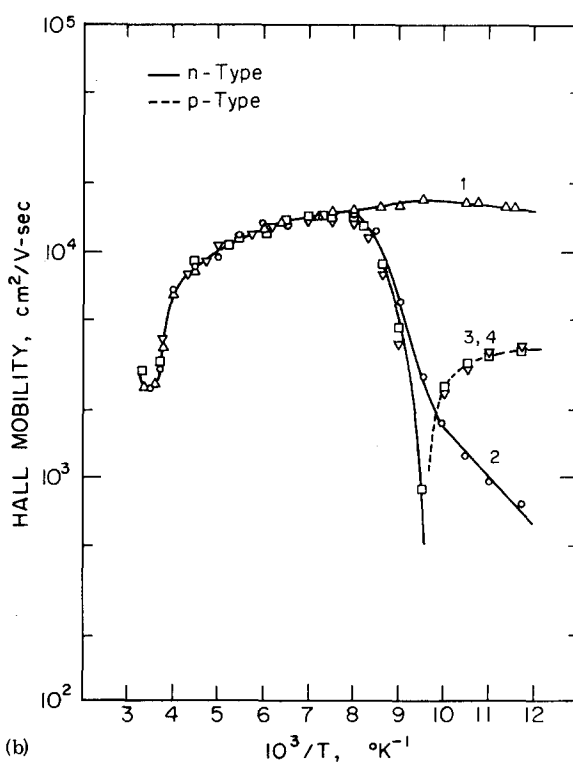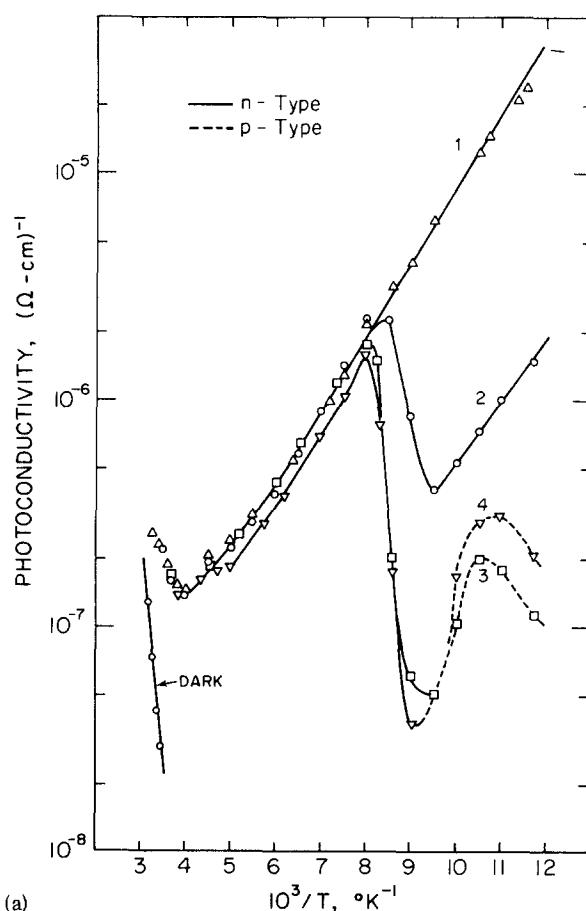

FIG. 4. (a) Temperature dependence of photoconductivity in GaAs :O for intrinsic photoexcitation after quenching 1 h with 1.08-eV (○) or 1.18-eV (□) photons at 82 °K, measured while warming; and for simultaneous intrinsic and 1.18-eV photoexcitation (▽). (b) Temperature dependence of the photo-Hall mobility for GaAs :O. Symbols are the same as in (a).

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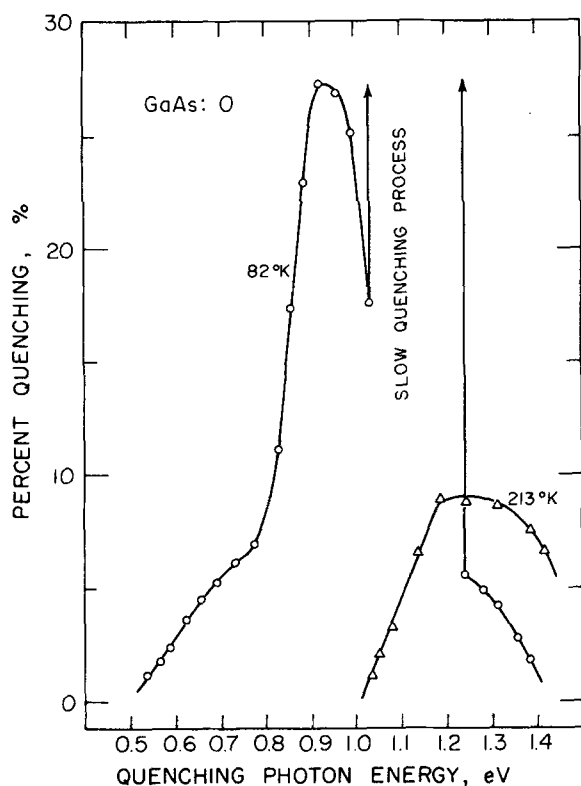
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FIG. 5. Optical quenching spectra for GaAs :O at 213 and 82 °K. Percent quenching is defined in terms of the difference between the maximum photocurrent obtained with both primary and quenching light on and the steady-state current under these conditions, divided by the maximum photocurrent, e.g., in the case that the quenching light produces no photoexcitation, the maximum photocurrent referred to is simply the photocurrent excited by the primary light.

Curves 2 and 3 represent the crystal in a low-sensitivity, at 82 °K. This quenching process is persistent at low temperatures, and the low-sensitivity quenched state remains unchanged long after the removal of the quenching radiation even with intrinsic photoexcitation present. Upon heating, however, a rapid and dramatic recovery from the low-sensitivity quenched state to the high-sensitivity state occurs when the temperature exceeds 105 °K and is complete by 125 °K. Curve 4 shows the temperature variation for simultaneous intrinsic photoexcitation and 1.18-eV irradiation during heating; except for the effects of a fast quenching process producing differences between 125 and 225 °K as discussed in Sec. II C, curves 3 and 4 are quite similar.

The excess photoexcited free-electron density is found to vary linearly with light intensity at different temperatures for incident flux densities of 10^{12}–10^{15} cm^{-2} sec^{-1}; a slightly superlinear behavior is observed below 10^{12} cm^{-2} sec^{-1} at 82 °K. Photo-Hall measurements show that the electron mobility is independent of photoexcitation to within 3% at each temperature, over a range of four orders of magnitude of excitation intensity.

C. Optical quenching of photoconductivity

The optical quenching spectra for photoconductivity at 82 and 213 °K are given in Fig. 5, i.e., below and

above the critical temperature for the persistent quenching effect. At 300 °K under the same experimental conditions there is no observable optical quenching over this entire spectral range.

At 213 °K, secondary light with photon energy between 0.7 and 1.0 eV increases the intrinsic-excited photoconductivity, indicating a level located at about 0.7 below the conduction band, the same level responsible for the low-energy threshold of the spectral response curves. For photon energies greater than 1.0 eV, the secondary light produces first an increase in photoconductivity and then a quenching. The quenching process indicates the existence of a level 1.0 eV above the valence band with a smaller electron cross section than the dominant recombination centers; quenching radiation excites electrons from the valence band to these levels at 1.0 eV and frees holes to participate in recombination with electrons.

At 82 °K, the onset of optical quenching is at about 0.5 eV with an abrupt increase at about 0.77 eV. For photon energies of the quenching light between 0.5 and 0.65 eV, and between 0.8 and 1.0 eV, only quenching is produced without accompanying excitation. Between 0.73 and 0.77 eV, there is a small excitation after rapid quenching; the quenching and recovery processes in these spectral regions are relatively fast. When the secondary photon energy exceeds 1.03 eV, however, three processes are found: (i) a fast excitation, (ii) a fast quenching, and (iii) a slow and very large quenching, corresponding to the persistent effects described above. At 1.08-eV quenching light, for example, the fast quenching produces 7.5% quenching; after 1 h of exposure, the slow quenching has produced 98% quenching with a decrease in the Hall mobility from 1.5×10^4 to 1.8×10^2 cm^2/V sec. If the quenching light is turned off after this exposure, leaving the intrinsic primary photoexcitation on, the time constant for recovery is very long. Even after a recovery period of 4.5 h at 82 °K, the photoconductivity recovers only 2% and the Hall mobility is 1.4×10^3 cm^2/V sec. At 1.15-eV quenching light, the photoconductivity converts from n to p type, and persists p type even several hours after turning off the quenching light.

D. Thermally stimulated conductivity

Measurements of thermally stimulated conductivity were made after photoexcitation by monochromatic intrinsic or extrinsic radiation. The results of thermally stimulated conductivity (TSC) and the corresponding Hall mobility after intrinsic photoexcitation with light intensity of 1.2×10^{15} cm^{-2} sec^{-1} for 30 min are shown in Fig. 6. The Hall mobility is n type and is about the same value as that of the intrinsic photo-Hall mobility given in Fig. 4(b), indicating that the measured TSC is contributed mainly from electron traps. Some eight electron traps are detectable,[20] most of which are common to a variety of high-resistivity GaAs samples.[13,21,22]

The electron capture cross section of each trap can be determined by measuring the kinetics of trap filling from TSC curves run after varying initial illumination periods. Estimated trap depths, densities, and capture
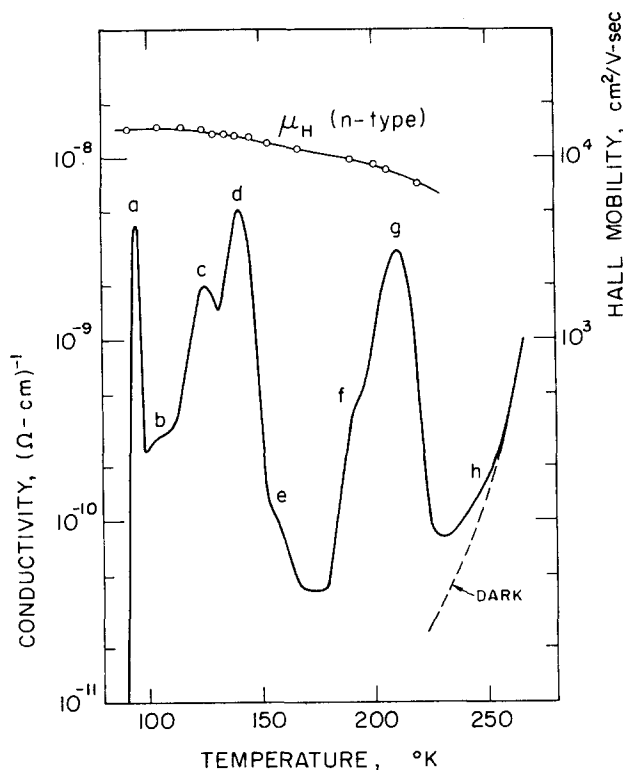
FIG. 6. Thermally stimulated conductivity and Hall mobility corresponding, as a function of temperature for GaAs :O.

cross sections are summarized in Table II. Traps a, c, and d (as labelled in Fig. 6) have an electron capture cross section almost 10^3 times smaller than trap g.

"Strange" properties of trap distributions after white-light excitation have been reported by Haisty,[22] in which certain TSC peaks grow and then decrease with increasing illumination time. Our work shows that this disappearance of shallow trap peaks with white-light illumination is a consequence of the quenching effect of photon energies in the range between 1.0 and 1.25 eV. The measured photocurrent and the TSC magnitudes for traps a, c, and d, for example, all increase to a maximum and then decrease for longer illumination time with 1.1-eV photons; trap g is unaffected since its peak temperature is above the critical temperature for thermal removal of the low-temperature-induced quenched state.

The electric field dependence of TSC curves was also investigated. Peaks a—e are independent of the applied field during photoexcitation (all TSC curves were measured with 10 V applied after photoexcitation) for electric fields up to about 60 V/cm, decrease slightly for fields between 60 and 200 V/cm, and decrease rapidly for fields above 200 V/cm. The magnitudes of traps f—h, however, are unaffected by the magnitude of the field applied during photoexcitation. The decrease in the magnitude of the TSC peaks a—e for an applied voltage of 700 V during excitation is by a factor of about 25.

E. Non-Ohmic electrical behavior

At 90 °K intrinsic photocurrents start to saturate at

about 60 V/cm and show oscillations with low frequency and small amplitude, as frequently reported in many semiconductors.[23–32] Generally it is believed that these oscillations are associated with the formation and motion of high-field domains. The domain velocity can be calculated from the oscillation period and the distance between electrodes. For GaAs :O, domain velocity decreases with increasing light intensity, and increases with increasing electric field over the field range 60—100 V/cm, and the light intensity range 10^{12}–10^{15} $cm^{-2} sec^{-1}$; domain velocities between 2×10^{-2} and 10 cm/sec were observed. When a field greater than 200 V/cm is applied to 90 °K, another type of oscillation occurs with larger oscillation amplitudes and lower frequencies. For these oscillations the domain velocity increases with light intensity, but decreases with electric field; velocity values between 10^{-2} and 3×10^{-1} cm/sec were observed. No oscillations are observed when the sample has been transformed to the low-sensitivity state by irradiation with photons with energy between 1.0 and 1.25 eV, and Ohmic electrical behavior is observed for fields up to 1400 V/cm.

III. DISCUSSION OF RESULTS

A. Energy level model for GaAs : O

To facilitate discussion of the results, an energy level model is proposed and shown in Fig. 7 which consistently describes the experimental observations. The numerical data given in Fig. 7 are for 82 °K and energy values have an uncertainty of ± 0.02 eV. Levels a—h are the electron traps summarized in Table II. Levels 1—4 are recombination centers. The model may be constructed as follows.

The temperature dependence of dark conductivity near room temperature is controlled by the thermal excitation of electrons from level 4 which lies 0.48 eV (at 0 °K; see Table I) below the conduction band. In thermal equilibrium, level 4 is mostly empty and levels 1—3 are mostly occupied. The low-energy photoconduc-

TABLE II. Properties of electron traps in GaAs :O.

Peak index	T_m	Trap density (cm^{-3})	Electron capture cross section (cm^2)	Trap depth (eV) [a]
a	95	$> 4 \times 10^{13}$	5×10^{-19}	0.15
b	111	$< 1 \times 10^{14}$	· · ·	0.19
c	126	5×10^{14}	7×10^{-19}	0.22
d	141	1×10^{15}	8×10^{-19}	0.25
e	156	$< 1 \times 10^{14}$	· · ·	0.28
f	191	$< 6 \times 10^{14}$	· · ·	0.35
g	212	4×10^{15}	3×10^{-16}	0.45
h	250	$< 1 \times 10^{14}$	· · ·	0.53

[a]Values of trap depth were measured for traps c, d, and g by the "decayed TSC" method; for the other traps they are a reasonable estimate based on a variety of ways of possibly determining the trap depth from the temperature of maximum TSC. In general, all the trap depths are approximately given by $E_t = 24k T_m$, traps a—f being somewhat less than this value, and traps g and h being somewhat greater than this value.

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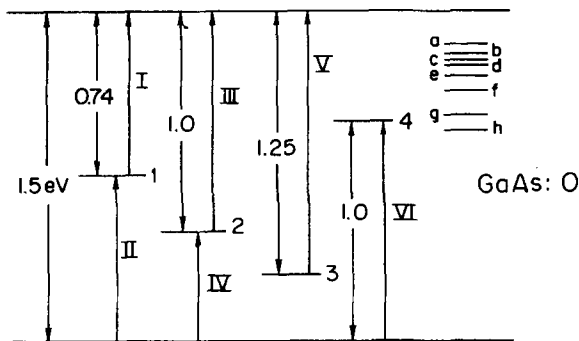
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FIG. 7. Proposed energy level scheme for the GaAs : O crystals of this investigation.

tivity threshold corresponds to transitions of type I, with a value of about 0.68 eV at 295 and 213 °K, and of about 0.74 eV at 82 °K. The slight structure at about 1.0 eV seen at 295 and 213 °K (Fig. 2) may indicate transitions of type III; as discussed in Sec. III B the slow persistent quenching process observed at low temperatures is also associated with transitions of type III. Transitions of type V are observed at 82 °K, corresponding to the rapid decrease of photoconductivity and photo-Hall mobility for photon energies less than 1.25 eV, when the spectral response curve was measured from high to low photon energies [Figs. 3(a) and 3(b)]. At 82 °K, the low-energy part of the optical quenching spectrum for photon energies less than 0.77 eV is due to both transitions of type IV and transitions from the valence band to level 3. The abrupt quenching process followed by a small excitation of photocurrent for photons between 0.73 and 0.77 eV corresponds to a competition between transitions of types I and IV. The sharp increase of quenching above 0.77 eV indicates that another quenching process corresponding to transitions of type II starts to compete and dominate. At 213 °K, the quenching process due to type-VI transitions is seen.

Since the energy location of level 4 deduced from the onset of the quenching spectrum at 213 °K (1.0 eV) is about the same as that of trap level g, which has a relatively large density, these two levels may be identical.

Level 1 is probably to be associated with oxygen impurity, since level 1 is the major imperfection level in our GaAs :O sample. In addition, a level near the middle of the band gap has been reported by many investigators for GaAs :O. If the level were associated with oxygen substituted on an As site, it should exhibit a donor character with $C_1^n > C_1^p$, where C_i^j is the capture coefficient of level i for carrier j, and the observed n-type intrinsic photoconductivity would be unlikely. If, however, the oxygen is in such a complex center that $C_1^n \sim C_1^p$, intrinsic photoconductivity is n type when the density of occupied centers is larger than the density of empty states. This is the condition for all of the samples measured in this investigation in the high-sensitivity state. Electrotransmission measurements indicate that transitions I are from a center with a dipole moment.[15]

B. Persistent quenching effect

The persistent quenching effect involves a reorganization of level occupancies that affects the electron-to-hole—lifetime ratio in a major way. Furthermore, once this reorganization is brought about by exposure to photons in the range between 1.0 and 1.25 eV, subsequent photoexcitation by intrinsic light cannot by itself reverse the process in a finite time. Raising the temperature above 105 °C, however, does reverse the process in a very short time. One mechanism that satisfies these requirements is the following.

Suppose that $C_2^n \gg C_1^n$, $C_2^n > C_2^p$, and $C_2^p \gtrsim C_1^p$ and $C_{t_i}^p$, where $C_{t_i}^p$ is the hole capture coefficient of the ith trapping level. Given these conditions, level 1 may still dominate the intrinsic photoconductivity if level 2 is originally completely occupied and $N_1 > N_2$, where N_1 and N_2 are the density of imperfection levels 1 and 2, respectively. The process of state transformation involves the transfer of electrons from level 2 to the traps via transition III at low temperatures. As level 2 is gradually emptied in this way, its role in recombination increases and the electron lifetime decreases; because of the large value of C_2^n, this emptying process is slow. At steady state in the low-sensitivity condition, both levels 2 and 1 are only partially occupied. After the low-sensitivity state is produced, it will persist at low temperature even under intrinsic photoexcitation. Because $C_2^p \gtrsim C_1^p$ and $C_{t_i}^p$, once a free electron is captured at level 2, it has a high probability of recombining with a hole, leaving level 2 unoccupied. On the other hand, the filling of level 2 and the reversal of the sensitivity transformation could be accelerated if more free electrons were available in the conduction band than free holes in the valence band; in short, if the electrons stored in traps were returned to level 2. Comparison of Figs. 4(a) and 4(b) with Fig. 6 and Table II shows that the temperature at which the rapid and dramatic recovery from the low-sensitivity to the high-sensitivity state occurs corresponds to the temperature at which the major trapping centers empty rapidly.

The initial assumption that $C_2^n \gg C_1^n$ does not necessarily contradict the observation in Fig. 5 of an onset for optical quenching at low temperature of 0.5 eV. This onset was measured at the limit of the monochromator used, where the light intensity was rapidly falling off with decreasing photon energy. It can be said only that the threshold for quenching is less than 0.5 eV, and probably corresponds to the transition from the valence band to level 3.

It is unlikely that level 2 is associated with simple impurity defects, but it is probably associated with a complex center also. If level 2 is associated with a donor-acceptor pair $(D^+A^-)^0$, for example, then the coefficient inequalities assumed above may be rationalized. The empty state of $(D^+A^-)^0$ is $(D^+A^0)^+$, which may be expected to have a very large cross section as the presence of the neighboring acceptor enhances the Coulomb cross section of the ionized donor, so that $C_2^n \gg C_1^n$. At the same time, it may be valid for $C_2^p \gtrsim C_1^p$ because a hole may be more easily captured by the neutral complex center $(D^+A^-)^0$ with its negative acceptor

than by a simple neutral center. That level 2 is a Coulomb attractive center is also indicated by the observation that for photoexcitation by 1.05 eV photons, the photocurrent increases faster than linearly with applied voltage for higher voltages.

A discussion of the identity of the electron trapping centers is deferred to the accompanying paper on GaAs:Cr.[13]

C. Photocurrent oscillations

Photocurrent saturation and low-frequency oscillations at high fields are found for photoexcitation with photons in excess of 1.3 eV. This effect can be ascribed to a field-dependent trapping effect similar to that first proposed for Au-doped Ge by Ridley and Watkins.[33] If level 3 were a Coulomb-repulsive center, its capture cross section would increase with electric field and the electron lifetime would decrease. Our previous identification of levels 1 and 2 indicates that their capture cross sections should be either independent of or decrease with electric field, and that therefore they are not responsible for the observed oscillations. Level 3 cannot be presently identified further because of a lack of information on the identity of double acceptor imperfections in GaAs.

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