Temperature dependence of the persistent photocurrent in Czochralski gallium arsenide

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The persistent-photocurrent effect in bulk, semi-insulating GaAs has been studied as a function of temperature. The persistent photocurrent is activated by illumination with 1.13-eV light and is preceded by an enhanced photocurrent during long-term illumination after the initial photocurrent has decayed in a quenching process similar to that of the deep donor *EL*2. Both the magnitude of the persistent photocurrent, and its decay rate are strongly temperature dependent and show significant reduction above 40 K. These results indicate that while the metastable transformation of *EL*2 is required for the activation of the persistent photocurrent, another unidentified metastable process after this is also required.

INTRODUCTION

Photoinduced reactions have provided a fertile field for the study of defects in semiconductors for many years. Semi-insulating (SI) GaAs has been studied extensively over these years because of its technological importance as a substrate for high-performance electronic devices and because of the wealth of fundamentally interesting effects observed, primarily due to the intrinsic deep donor *EL*2,[1] which undergoes a metastable transformation at low temperatures when illuminated with light in a band centered near 1.1 eV. Recently, a persistent photocurrent (PPC) has been observed in SI GaAs.[2–6] Since the transformation of *EL*2 into its metastable state, denoted *EL*2*, results in the disappearance of such effects as infrared absorption, photocapacitance, and photoluminescence,[1] the photoinduced increase in a parameter such as the photocurrent is unusual.

Jimenez *et al.*[2–5] reported an unexpected enhancement in the photocurrent (PC) of SI GaAs at 80 K under 1.3-eV illumination in some of their samples. In these samples the PC initially decayed, as expected for photoionization of *EL*2 undergoing photoquenching, but after a time the PC started to increase until it saturated at a value exceeding the initial value of the PC. The current remained at high values long after the illumination was removed. This dark current is the PPC. We will refer to the saturated photocurrent as the enhanced photocurrent (EPC). Prior to the work of Jimenez *et al.*, most PC experiments on bulk GaAs had shown that the PC quenches under sub-band-gap illumination.[7] After the PC experiments, Wan and Bray[8] observed electronic excited-state transitions from neutral carbon and zinc at 15 K in previously SI GaAs, after quenching *EL*2, by electronic Raman spectroscopy (ERS). The results of Wan and Bray were corroborated by the infrared-absorption experiments of Wagner *et al.*,[9] who observed the excited-state absorption spectra from the neutral impurities only after quenching. Fuchs and Dischler,[10] in a different absorption experiment, have reported the presence of free holes after the quenching of *EL*2. These optical experiments suggest that the PPC observed by Jimenez *et al.* and Mitchel *et al.*[6] is due to thermal ionization of neutral acceptors, predominantly carbon and zinc, which are normally compensated by *EL*2.

Here we report further experiments on the PPC effect in SI GaAs, including temporal and temperature-dependent effects. We show that the PPC at temperatures below 40 K is most likely due to thermal ionization of shallow acceptors such as carbon. Above 40 K the PPC decreases rapidly with both time and temperature in a manner more similar to detrapping than thermal activation of shallow levels. A two-stage process is required to activate the PPC, and the first stage is the normal metastable transformation of *EL*2 into *EL*2*. The charge transfer that leaves the shallow acceptors neutral occurs during the second stage, which is unidentified at this time.

The experimental details including samples and measurement techniques will be described in the next section, followed by the experimental results. These and previous results will be discussed in the fourth section.

EXPERIMENTAL DETAILS

The samples used in this study came from several different laboratories and included both Czochralski- and Bridgman-grown material. All were semi-insulating and were either undoped, very lightly oxygen doped (Ga_2O_3), or indium alloyed. Carbon concentration as determined by local-vibrational-mode infrared absorption varied from below the detectability limit of 10^{14} cm^{-3} to 3×10^{16} cm^{-3}. *EL*2 concentrations, also determined by infrared absorption, varied between 5×10^{15} and 50×10^{15} cm^{-3}. Samples in the van der Pauw configuration (either squares or crosses) were cut from wafers. Since both the EPC and PPC in these materials were found to be *p* type, Ohmic contacts were made by alloying In–5% Zn solder at 425 °C for 2 min. While the contacts were not Ohmic during the initial stages of the PC quench, after the EPC formed they were quite Ohmic. The initial photocurrent in SI GaAs is *n* type, so this behavior is to be expected for *p*-type contacts.

The experiments were performed in a variable-

temperature helium-vapor cryostat. The sample current and voltage were measured and recorded by a personal computer. Current was measured on a Keithley 619 digital electrometer. The temperature was measured with a calibrated silicon-diode thermometer. Illumination for the photocurrent studies was provided by a quartz halogen lamp and narrow-bandpass filter centered at 1.13 eV (full width at half maximum 0.01 eV). The lamp was kept on and the sample illuminated by a computer-controlled shutter on the cryostat. For most temperature-dependence studies, power was applied to the sample mount through a resistive heater, and the temperature was allowed to drift up slowly as current and voltage values were recorded. No attempt was made to ensure that the heating rate was constant. It took about 2 h to sweep from 8 to 150 K.

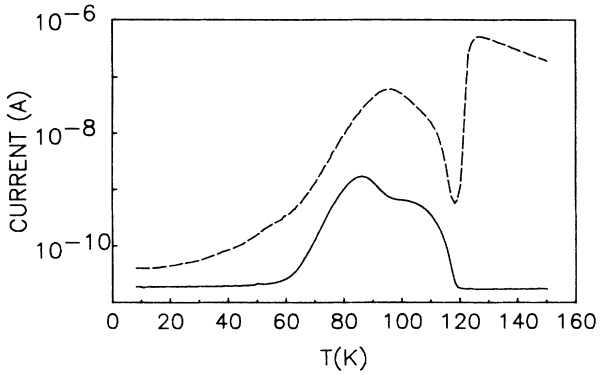

FIG. 2. Temperature dependence of the 1.1-eV photocurrent and the dark current, both after illumination at 8 K, for a sample with normal quenching behavior, — — —, photocurrent; ——, dark current.

RESULTS

While a variety of effects were observed in low-temperature PC versus time experiments, most samples either showed "normal" photoquenching of the PC, which is represented in Fig. 1 for a Czochralski sample at 8 K, or showed an enhanced photocurrent (EPC) which developed after the initial photocurrent decreased to near the detection limit. The EPC was accompanied by a persistent photocurrent (PPC) after the illumination was removed. Normal quenching of the PC resembles the quenching of such other properties as the photocapacitance and the infrared absorption, which are most directly associated with $EL2$.[1] The decay of the PC in normal quenching is nearly exponential until the lowest currents, where saturation is observed. The PC remained at or near 10^{-10} A for hours with no observable change. Photo-Hall experiments at 80 K showed that the initial PC is n type and that the PC in the tail is p type. The temperature dependence of the photocurrent and dark current after quenching at 8 K are shown in Fig. 2. Here the data were recorded as the temperature was allowed to sweep up to 150 K from 8 K after the sample had been illuminated for 45 min or more. The peaks in the dark current near 80 and 100 K are similar to the thermally-stimulated-current (TSC) peaks reported by others,[11–13] and are most likely due to detrapping effects. The dip at 130 K in the photocurrent is the conversion from p- to n-type conduction that occurs when $EL2$ recovers from its metastable state. All Bridgman-grown samples, including both horizontal and vertical grown crystals, and samples from both quartz and boron nitride boats, showed this normal quenching[14] as well as a small number of Czochralski-grown samples. Quenching experiments at higher temperatures resulted in similar effects for these samples up to temperatures exceeding 100 K. Only in the range above 70 K was there a residual PC due to the TSC-like peaks. This did not result in a PPC.[15]

The enhanced photocurrent (EPC) and persistent photocurrent (PPC) comprise the dominant behavior exhibited by undoped Czochralski samples including In-alloyed material. These effects are shown in Fig. 3, which is the time dependence of the 1.1-eV PC and PPC for an undoped, unalloyed Czochralski sample. Initially the PC is very similar to the normal quenching effect in Fig. 1, but immediately after the current decays to the lowest value, which is usually in the same range as the residual current of the normal samples, it starts to rise at a rate close to but not identical to that at which it initially quenched until it slows down and saturates at a current usually con-

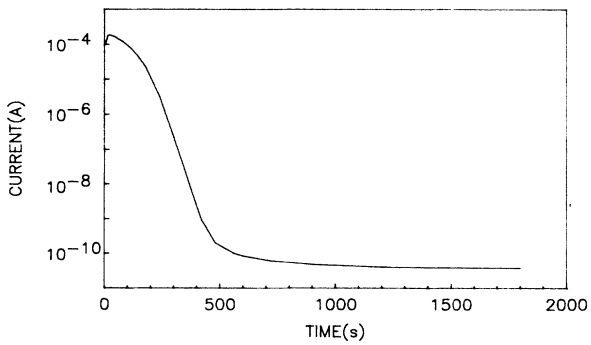

FIG. 1. Normal photoquenching of 1.1-eV photocurrent at 8 K for a Czochralski-grown sample.

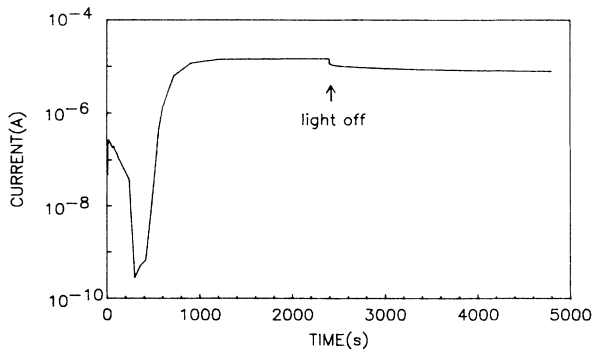

FIG. 3. 1.1-eV enhanced photocurrent and persistent photocurrent at 8 K.

siderably higher than the initial peak. In only two of the samples we studied did we observe a delay prior to the onset of the enhanced photocurrent similar to that reported by Jimenez *et al.*[2-4] In these samples, the PC remained in the 10^{-10}-A range for several minutes after the initial quench before starting to increase. In all other samples showing an EPC the increase followed immediately after the decay with no delay, as shown in Fig. 3. Once the illumination was removed, the current first dropped sharply and then EPC decayed very slowly. The dark current remained within an order of magnitude of the EPC for hours. Both the EPC and PPC were *p* type, as determined by Hall-effect experiments.

The temperature dependences of the EPC and PPC were determined by sweeping the temperature after saturation was achieved, either under illumination or in the dark, respectively. The PPC was allowed to decay for several minutes prior to heating. These results are shown in Fig. 4. In the figure the temperature dependencies of the EPC and PPC are compared to the dark current of an undoped *p*-type sample that was conducting at low temperatures due to thermal ionization from uncompensated acceptors. This conducting sample was dominated by a carbon and the (77, 203 meV) double acceptor common to undoped Czochralski material grown from gallium-rich melts.[16] The EPC and PPC are nearly identical at the lowest temperatures and are very similar to the *p*-type sample as well. This suggests that both the EPC and PPC are predominantly due to thermal ionization of shallow acceptors. As the temperature is increased, both the EPC and the PPC fall off, with the PPC decaying rapidly at temperatures above 50 K in this sample. In general, the PPC started to decay in the range 40–60 K with some sample-to-sample variation, and the EPC stopped increasing with temperature in the same range. The dip in the EPC near 120 K is reminiscent of that in Fig. 2 and is most likely due to the recovery of *EL*2 from its metastable state. It should be noted that all of the PPC is annealed well before the dip in the EPC. The *p*-type sample in Fig. 4 was taken from the tail of a Czochralski boule that converted from *n*-type semi-insulating at the

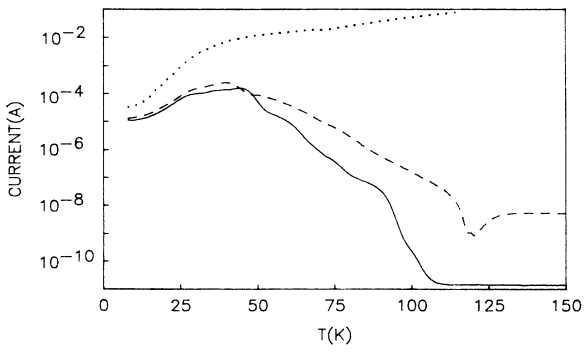

FIG. 4. Temperature dependence of the 1.1-eV photocurrent (EPC) and the dark current (PPC) after illumination at 8 K compared with the dark current of an undoped, *p*-type sample dominated by carbon and the intrinsic double acceptor. — — —, EPC; ——, PPC; · · ·, dark current of *p*-type sample.

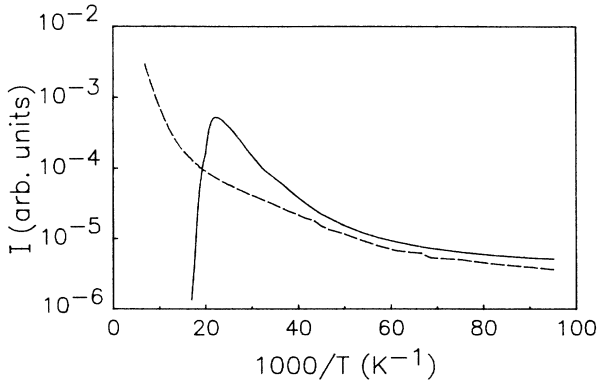

FIG. 5. Scaled current vs temperature for a semi-insulating seed sample and *p*-type tail sample from the same boule. The PPC for the seed sample was activated by illumination with 1.1-eV light. No illumination was used on the *p*-type sample. ——, seed; — — —, tail.

seed end to *p*-type conducting at the tail due to arsenic loss during growth. The SI seed-end samples exhibited EPC and/or PPC effects similar to Figs. 3 and 4. The seed and tail samples from this boule are compared in Fig. 5, where we plot the PPC after activation with 1.1-eV light for the seed sample and the dark current of the tail sample without any prior illumination versus inverse temperature. The currents are scaled in arbitrary units because different applied voltages were used for the two samples. At the lowest temperatures the slopes of the two samples are nearly identical, supporting the hypothesis that the PPC is due to thermal activation of neutral carbon. The steeper slope for the seed sample in the range 24–45 K could be due to the presence of a deeper acceptor. Since this slope is shallower than that in the tail sample due to the 77-meV level of the double acceptor at the highest temperatures shown, it is unlikely that this unknown acceptor in the seed sample is the double acceptor. The slope of the PPC versus inverse temperature in this range varied considerably between samples, and no general trend could be determined, indicating perhaps that more than one level could be present.

Time-dependent quenching experiments were performed at different temperatures to study the activation and decay process. The saturation current of the EPC was observed to vary in a manner similar to the temperature dependence of the photocurrent in Fig. 4; that is, it increased up to about 40 K and then fell significantly at higher temperatures. The EPC at saturation was almost identical to the value of the photocurrent measured in the temperature sweep at the particular temperature. The decay rate of the PPC was found to depend strongly on temperature as well. The normalized PPC versus time for several temperatures is shown in Fig. 6. The change from a relatively slow decay at low temperatures to a much more rapid decay at higher temperatures occurs near 40 K. The annealing process was found to be complex. Complete removal of the EPC and PPC was achieved by heating to about 135 K. If, after heating the sample to this temperature, it was then cooled in the dark

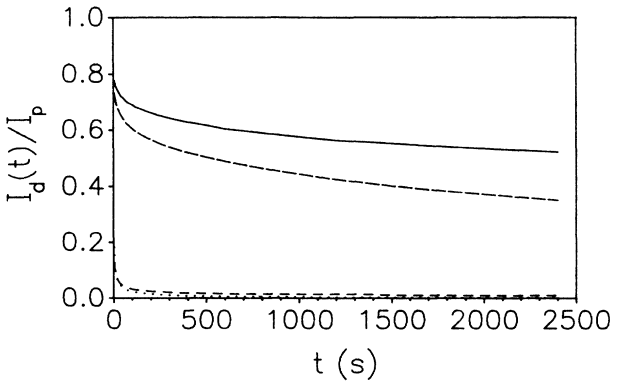

FIG. 6. Temperature dependence of the decay rate of the PPC. $I_d(t)/I_p$ vs time after quenching with 1.1-eV light at various temperatures. I_d=PPC, I_p=EPC. ——, 8 K; — — —, 20 K; —·—·—·, 40 K; · · ·, 80 K.

back to 8 K, no PPC was measurable and the PC versus time dependence was identical to the original activation when the sample was reilluminated. The 8-K PPC could be completely removed by heating the sample to temperatures as low as 45 K and then cooling back to 8 K, but in this case, upon reactivation, the PC went immediately to the saturation value, with only a rise time on the order of 1 or 2 min and no structure such as seen in Fig. 3 was present. While the 8-K PPC could be removed by heating, if the sample was heated again in the dark without reactivation the higher-temperature PPC would return once the anealing temperature was reached and the current would trace out the rest of the curve seen in Fig. 4. This only occurred above 40 K. Heating to temperatures lower than this did not significantly affect the 8-K PPC; only a slight reduction in the PPC attributable to slow decay was observed. The reproducibility of the lowest-temperature PPC is indicative of thermally activated current, while the behavior of the PPC above 40 K is more similar to detrapping effects.

Finally, we note that none of the SI Czochralski samples that were oxygen doped or whose deep-center photoluminescence was dominated by the 0.63-eV *ELO* emission as opposed to the 0.68-eV *EL*2 emission[17] showed any EPC or PPC. The presence of large amounts of *ELO* thus appears to suppress these effects.

DISCUSSION

The PPC reported here is almost certainly due to the same process that results in neutral shallow acceptors observed in optical experiments.[8–10] The activating illumination is similar [the optical experiments used the 1.17-eV line of a neodymium-doped yttrium aluminum garnet (Nd:YAG) laser] and the temperature dependence of the PPC below 40 K is consistent with thermal activation from neutral acceptors like carbon, which was detected by the optical experiments as well. Fuchs and Dischler,[10] in infrared-absorption experiments, indicated that the free holes they detected after quenching *EL*2 in SI Czochralski samples annealed in the same range as where we observe the onset of PPC decay, 20–50 K in their experiment. We will therefore assume that the activation processes for the electrical and optical effects are identical. Since the low-temperature EPC and PPC are nearly identical, we will further assume that they are both due primarily to thermal activation of neutral acceptors and that photoionization processes in the EPC are not significant to the effects reported here. The fact that both the EPC and PPC are *p* type and thermally activated almost certainly indicates that the conduction is due to thermal ionization of shallow acceptors. The observation of neutral carbon and zinc by the optical experiments, the fact that these are the most common impurities in bulk GaAs, and the similarity of the current-versus-temperature curves for unquenched *p*-type material and quenched SI material suggests that the EPC and/or PPC are (is), in fact, due primarily to carbon and zinc with other acceptors, either impurities or defects, possibly contributing to the current as well. However, the conclusion that carbon and zinc are dominant will not be verified until a complete analysis of the temperature dependence of the EPC and/or PPC hole concentration is performed to identify the activation energy. For the rest of the discussion we will assume that the EPC and/or PPC are (is) due to thermal activation of shallow acceptors, but leave the identification of these for later experiments.

For initially compensated acceptors to become neutral, there must be a transfer of electrons from the acceptors. The electronic structure of the acceptors does not appear to be different in normal *p*-type material and in the PPC state. This can be shown by the fact that the electronic excited-state spectra are identical as determined by both ERS (Ref. 8) and infrared absorption.[9] This suggests that the shallow acceptors themselves do not undergo a metastable transformation because it is hard to imagine any defect transformation that would result in a charge transfer from a defect that leaves the fine-line excited-state spectra unchanged and unshifted. We therefore conclude that the acceptors are passive recipients of some defect process that affects the charge state of the compensating deep donors, *EL*2 in particular. This defect process is not, however, the usual *EL*2 metastable transition, *EL*2 to *EL*2*, since the temperature dependence of the PPC is incompatible with the temperature dependence of the activation of and recovery from *EL*2*. The PPC has a noticeable decay, even at 8 K, where the recovery of *EL*2 is almost negligible. The temperature dependence of the decay rate is not compatible with the thermal recovery of *EL*2.[7] Infrared-absorption studies of *EL*2 in thick samples from the same boules as some of our PC samples show that *EL*2 remains in its metastable state after the PPC has been removed by 80-K annealing. It does appear, however, that although the *EL*2 metastable transition can not completely explain the PPC effect, it is most likely necessary before the EPC and PPC can be activated. The initial quenching of the PC strongly resembles that observed in samples that do not show a PPC, which suggests they are due to the same process which we attribute to the quenching of *EL*2. The fact that the activation of the EPC and/or PPC is different after an-

nealing at 80 K than at 135 K or higher indicates that two separate processes are required. We proposed that the first is the quenching of $EL2$ into $EL2^*$ and that the second is the process that results in the charge transfer from the shallow acceptors. We are not prepared at this time to support a process that would result in the required charge transfer, but two general possibilities exist. First, the electronic level of the metastable, compensating deep donor, $EL2^*$, could be moved during the second metastable transformation to a new position below the level of the shallow acceptors, resulting in a defect in or near the valence band and uncompensated acceptors. This is essentially the process proposed by Dabrowski and Scheffler in a theoretical analysis of metastability of the isolated arsenic antisite.[18] Second, photoionization of the acceptors to the metastable defect could result in trapping of the electrons on the deeper defect. The rather complex recovery process seen in Fig. 4 does suggest a detrapping-like process. Further studies of the annealing of the PPC are required to identify which, if either, of these possibilities is correct.

The nature of the second process is also a mystery at this time, but it almost certainly must result in a change in $EL2^*$, since this defect state appears to compensate acceptors in samples that do not show the EPC and/or PPC effects. Jimenez *et al.*[4,5] have suggested that the defect $EL6$ (Ref. 19) is responsible and might be the defect that undergoes the metastable transformation resulting in the EPC and/or PPC. Our results indicate that $EL6$ is probably not the defect responsible for the effects. Among the SI Bridgman samples, from one of the suppliers we received several undoped conducting samples grown in the same reactor under near identical conditions as the SI samples. Deep-level transient-spectroscopy (DLTS) measurements were performed on the conducting samples and, in addition to $EL2$, the dominant deep level was found to be $EL6$. It seems reasonable that $EL6$ would also be present in the SI samples. Fang *et al.*[19] have indicated that $EL6$ is a dominant defect in Bridgman material, while our results indicate that the EPC and/or PPC effects are absent in undoped Bridgman-grown material.

In recent years several as yet unidentified electron-paramagnetic-resonance (EPR) spectra have been reported in GaAs that have photoactive properties similar to some of the effects reported here. The $FR1$, $FR2$, and $FR3$ (Refs. 20–23) and the $BE1$ (Ref. 24) signals appear only after sub-band-gap illumination at low temperatures. The $FR1$ (Ref. 20) signal appears after the $EL2$-related EPR signal is quenched and $FR1$ is annealed at 80 K, after which the $FR2$ signal appears.[23] The $FR3$ (Ref. 22) $BE1$ (Ref. 24) signals also appear after the $EL2$-related signal is quenched. $BE1$ anneals at 90 K, but we were unable to find an annealing temperature for $FR3$ in the literature. Kaufmann *et al.*[22] reported that the $FR3$ signal decayed slightly after the illumination was removed, which is similar to the EPC and/or PPC, and that it was only detected in Czochralski material. They concluded that the photoactive behavior was not due to any changes in the defect responsible for the $FR3$ signal, but rather to the quenching of $EL2$. Further studies of the spectral dependence of the EPC and/or PPC effects are needed to determine if the EPR defects are related to the EPC and/or PPC.

The question of why normal quenching is universally observed in undoped Bridgman material but is seen in only a small percentage of the undoped Czochralski samples deserves discussion. We studied several Bridgman samples from three different sources who used significantly different techniques, two of which were horizontal and the other vertical. In none of these samples was an EPC or PPC detected.[14] Of the many undoped Czochralski boules studied, only two showed exhibited unequivocal normal quenching. One of these boules was indium alloyed and was grown in a reduced thermal-gradient crystal puller. We were not given any details on the growth of the other sample other than that it was undoped and Czochralski grown. Since the indium-alloyed crystal had a very low dislocation density of a few thousand dislocations per square centimeter, we at first suspected that these defects might be responsible for the EPC and/or PPC effects, since Bridgman material usually has a dislocation density of a few thousand dislocations per square centimeter or less as well, while standard Czochralski material has densities greater than 10^4 cm^{-2}. However, another indium-alloyed boule grown in the same puller under identical conditions as the first showed EPC and/or PPC effects similar to Figs. 3 and 4. Nonetheless, the thermal gradients during growth are significantly lower in the Bridgman process than in the Czochralski process. The lower gradients in Bridgman growth might well result in less complex intrinsic defects than in Czochralski growth, such as isolated antisites instead of more complex defects. Another difference between the Czochralski and Bridgman techniques is the presence of the boric oxide encapsulant in the Czochralski technique, which leads to significant boron concentrations in the crystals. It may be noted as well that it has been suggested that $EL2$ is not a unique defect, but rather a family of similar defects with differences in the photocapacitance quenching for samples grown by different techniques.[25] It is conceivable that one member of the family shows EPC and/or PPC and another does not.

The absence of the EPC and/or PPC effects in oxygen-doped samples and samples whose deep-level PL is dominated by ELO, a deep donor with an ionization energy very close to that of $EL2$, suggests that this level does not participate in the second metastable transformation. If ELO is the dominant deep donor in these samples, then it would be the compensating level; thus any $EL2$ could undergo the double transformation, but this would not be detected because the ELO would still compensate for the acceptors.

CONCLUSIONS

We have observed an enhanced photocurrent and a persistent photocurrent in many Czochralski-grown undoped semi-insulating GaAs samples induced by illumination with 1.1-eV light at low temperatures. The activating process was found to be a two-stage process, the first stage being the normal quenching of $EL2$ and the second stage resulting in a charge transfer from previ-

ously compensated shallow acceptors, most likely by a reduction in the energy of the $EL2^*$ donor level. Both the EPC and PPC are p-type conducting, most likely due to thermal activation from shallow acceptors. The PPC decay rate was found to increase significantly at temperatures at or above 40 K. The EPC and/or PPC effects were observed in both unalloyed and indium-alloyed material. The effects were completely absent in undoped SI Bridgman samples and samples dominated by $EL0$ photoluminescence.

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