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Optically induced excess hole population in semi-insulating GaAs

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The existence of a persistent hole population in semi-insulating GaAs is studied in relation to both the photoquenching and enhanced photocurrent effects, which are achieved by persistent illumination with 1–1.35-eV light at low temperature. The observation of this hole population is related to the metastability of the *EL*2 level for the photoquenching, and to another state tentatively related to the *EL*6 level for the enhanced photocurrent. The role of the acceptors is discussed for both effects, being the enhanced photocurrent associated with the thermal emission of holes from the acceptors arising from the metastability. The persistent photocurrent observed in liquid-encapsulated Czochralski grown specimens is also discussed in terms of thermal emission from these acceptors.

I. INTRODUCTION

Recently, a controversy has arisen about the origin of the persistent hole population, either free or bound, observed after prolonged excitation of semi-insulating GaAs with light of the 1–1.3-eV spectral range at low temperature ($T < 140$ K).[1–6] This is an important point of discussion because the existence of such a hole population is usually related to the optically induced metastability of the so-called *EL*2 level;[7] on the other hand, it stretches the role played by the shallow acceptors in such an important and intriguing defect.[8] Besides, an understanding of how the acceptor levels providing these holes participate in the metastability of the *EL*2 level can help clarify the important problem of the charge state of both *EL*2 and *EL*2* defects. This is one of the keys together with the assessment of the electrical activity of the metastable state for the identification of the *EL*2 level. The existence of such an excess hole population can be observed by various experimental means and presents different characteristics depending on the samples, the temperature, and the optical excitation (wavelength, photon, flux, etc.) inducing the metastability. Photo-Hall,[1] electronic Raman scattering,[2] and free-carrier absorption[6] have shown the existence of such a hole population, either free or bound.

Time-resolved photocurrent experiments have demonstrated the existence of two consecutive induced effects by persistent excitation with light in the 1–1.3 eV spectral range: (i) photoquenching (PQ) and (ii) enhanced photocurrent (EPC).

Both have been extensively studied before,[1,6,9,10] and have been ascribed to the existence of lattice relaxations leading to metastable configurations of the defects displaying these effects; the photoquenching is due to the lattice relaxation of the *EL*2, while the EPC is tentatively ascribed to a metastability related to the *EL*6 level.

In the following sections the relation of these effects to the excess hole population will be discussed.

II. EXPERIMENT AND SAMPLES

Photocurrent experiments were carried out in semi-insulating (10^7–10^9 Ω cm) GaAs samples cut from either LEC (liquid-encapsulated Czochralski) or HB (horizontal Bridgman) ingots. The LEC material was undoped, although some In-doped samples were also studied. The HE samples were doped with low concentrations of chromium, and a small amount of Ga_2O_3 was added to the melt; all of the samples were semi-insulating *n* type. Hall measurements showed that *EL*2 pins the Fermi level at the midgap. Typically, *EL*2 concentrations were found to lie around 10^{16} cm^{-3} by the absorption calibration method by Martín.[11] The samples were tailored in $5\times2\times0.3$-mm^3 parallelepipeds. Electrical contacts were made using either silver, indium, or gold; no differences were found among them, when the measurements were carried out at low bias, in the I_{ph} versus V linear plot region.

The samples were mounted in the cold finger of a closed-helium-circuit Air Products cryostat, which allowed us to work from 10 up to 300 K. For thermally stimulated current (TSC) experiments the sample was mounted in a special holder, equipped with a light pipe and electric contacts, which was directly immersed in a liquid-helium bottle, which guarantees the absence of background infrared radiation.

Illumination was provided by light from a 150-W halogen lamp passing through a high-luminosity Bausch-Lomb grating monochromator. A filter system prevented high-order wavelength light. A 1-W cw neodymium-doped yttrium-aluminum-garnet (YAG) laser was used to produce the metastable transformation in the TSC and I_{ph} versus T experiments. The electric current was measured with either a Keithley 610 electrometer or a Keithley 26000 logarithmic picoammeter.

III. RESULTS AND DISCUSSION

The photoquenching (PQ) is unambiguously related to the metastability of the $EL2$ level, while the enhanced photocurrent (EPC) has been tentatively associated in our previous works[6,12] with a metastable transformation of the so-called $EL6$ level, of which the chemical nature is unknown. This level is ubiquitous in bulk GaAs and is usually identified as a deep donor ($E_c - 0.35$ eV); its concentration is similar to that of the $EL2$ level, around a few 10^{16} cm^{-3}. On the other hand its photoionization spectrum has been reported to be similar to that of the $EL2$ level,[13] which supports the hypothesis that the $EL6$ level also involves the As_{Ga} defect. Each phenomenon coexists in most of the samples studied. Usually, the rate of introduction of each phenomenon differs. The photoquenching takes place earlier, while the enhanced photocurrent is induced by further illumination. Figure 1 shows a typical I_{ph}(1.1 eV) versus t plot, in a sample exhibiting both effects. Figure 2 shows the rate of introduction of both effects as a function of the incident photon flux. Figure 3 evidences the influence of the temperature on the I_{ph}(1.1 eV) versus t plot. An adequate choice of the light intensity and wavelength and the temperature of the excitation has allowed us to separate the observation of these phenomena. However, because both of them are strongly sample dependent, such a separation is not always possible, and there exist specimens for which only the enhanced photocurrent was observed. Presumably it overshadows the observation of the photoquenching.

Taking these considerations into account, it is important to study each of the effects separately in order to gain insight into the origin of the excess hole population, which are the dominant carriers when both the PQ and the EPC states have been optically induced. The existence of this hole population constitutes one of the more intriguing aspects of the compensation of GaAs once the metastable state has been reached.

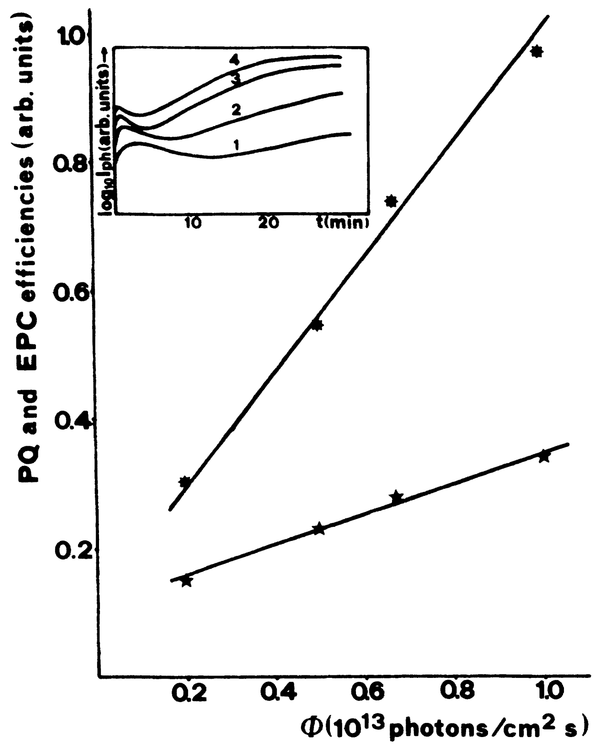

FIG. 2. Rate of introduction of the PQ and EPC, respectively as a function of the 1.1-eV incident photon flux. The inset shows the corresponding I_{ph} vs t plots.

A. Photoquenching

The main features of this phenomenon are as follows.

(1) This effect is observed in photocurrent experiments

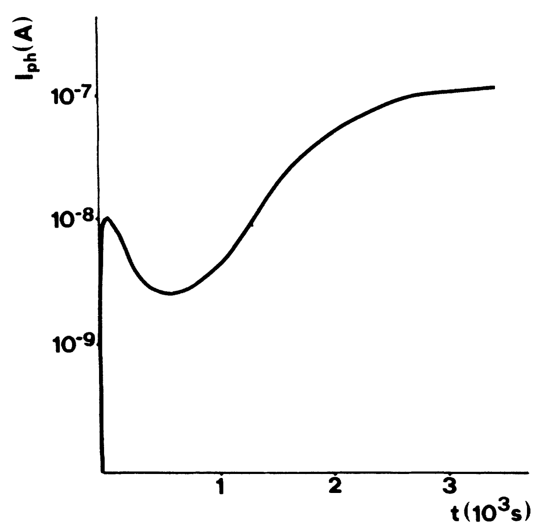

FIG. 1. I_{ph} vs the time of illumination with 1.1-eV light at liquid-nitrogen temperature for a LEC sample.

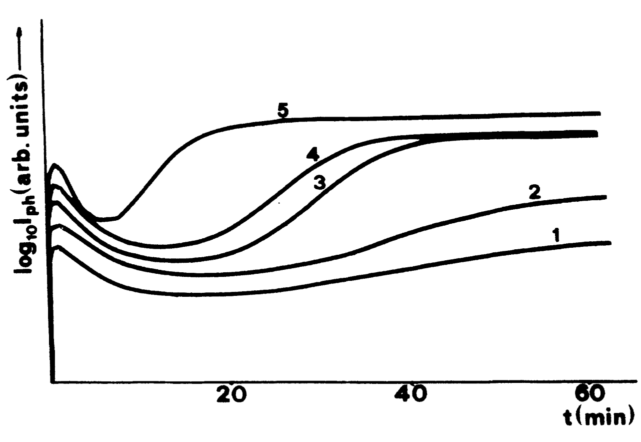

FIG. 3. Influence of the temperature on the I_{ph} (1.1 eV) vs t plots; (1) 25, (2) 35, (3) 50, (4) 60, and (5) 80 K.

as a continuous decrease of the photocurrent intensity.

(2) The photocurrent can be thermally recovered by heating above 140 K in darkness.

(3) It is partially recovered by exciting with either 0.9- or 1.45-eV light.

(4) A delayed photoquenching of the 1.45-eV photocurrent is also observed for further illumination with quenching light (1–1.3 eV).[14]

The *p*-type dominant conduction observed when *EL*2 has been transformed into the metastable configuration has been explained on the basis of a noncompensation of the residual shallow acceptors, mainly C and Zn, by the sequential transformation $EL2^{+} \rightarrow EL2^{0} \rightarrow EL2^{*0}$.[2,15]

It should be noted that in darkness and prior to any excitation the Fermi level is pinned at the (0/+) ionization level of the *EL*2 level, thereby compensating the residual acceptors. Nevertheless, this pure electronic mechanism presents some inconsistencies. The first one is that the free-hole concentration, estimated by photo-Hall measurements in the photoquenched state at 80 K, when all the shallow acceptors are ionized, lies around 10^9 cm^{-3},[1,16] which is several orders of magnitude below that expected from the lack of compensation of the residual shallow acceptors ($\sim 10^{15}$–10^{16} cm^{-3}). This seems to imply two possibilities. The first is that $EL2^*$ has donor levels in the band gap that compensate the residual acceptors; in other words $EL2^*$ is not neutral as is usually believed. The other possibility is the existence of other deep acceptor levels that trap the free holes arising from the uncompensated residual acceptors.

Nevertheless, the role of the residual acceptors is complicated by the quenching observed in the near-band-gap optical absorption and photocurrent. As has been described elsewhere, excitation with quenching light results not only in the quenching of the *EL*2 absorption band, but also near the band gap ($h\nu > 1.42$ eV) the absorption and the photocurrent are quenched. These observations suggest that the optical transitions arising from levels lying less than 120 meV above the top of the valence band are quenched; therefore, the acceptors initially pinned at such levels must necessarily shift to deeper positions in the band gap. This is confirmed by TSC experiments carried out in quenching conditions in the low-temperature region $T < 80$ K.[17] Compared with the unquenched TSC curve, the trap population is not only significantly quenched, but the temperatures at which the TSC peaks are observed shift apart from each other. Taking this into account, we can assume that the metastable state must involve not only the deep levels associated with *EL*2, but also other levels that, when unquenched, behave as shallow acceptors—less than 120 meV above the top of the valence band—but once the change in the defect configuration has occurred do not work this way, as is inferred from the absence of the optical and thermal activity related to them. The existence of such acceptors seems to support compensation models involving not only the shallow donors and acceptors and *EL*2, but also the acceptors, which must be considered in such a compensation model. On the other hand, if these acceptors participate in the metastability they must be related to the *EL*2 defect.

This seems to be reasonable, at least for the advanced stages of the quenching, for which the quenching of the near-band-gap photocurrent is observed.[18,19] Recently we have shown that once the 1–1.3-eV photocurrent quenching has been produced, further excitation with quenching light (1–1.3 eV) induces a quenching of the near intrinsic photocurrent ($h\nu > 1.42$ eV). It should be noted that because of the nonsimultaneity of these photoquenching effects (1.1 and 1.45 eV), the persistent hole population can hardly be justified as being the only consequence of the lack of compensation of acceptors due to the successive transformations undergone by *EL*2 when excited with 1–1.3-eV light. In this case it can be argued that a lattice relaxation, operating on the metastable state itself, which is monitored by the observation of the quenching of the near intrinsic photocurrent, is needed to account for this observation; in other words, a second metastable state appears in which the acceptor levels participate. These acceptor levels have to be pinned to the $EL2^*$ levels. That is to say, they are transformed by the metastability of *EL*2, which seems to indicate a rather complex nature of the *EL*2 atomic configuration, at least in the metastable state.

B. Enhanced photocurrent

The second effect showing the existence of an excess hole population is the enhanced photocurrent (EPC),[1,3,9,10,16] which we have tentatively associated with the so-called *EL*6 level.[5,12] This effect is observed under the influence of persistent excitation with 1–1.35 eV light at low temperature ($T < 140$ K); its metastable nature has been largely discussed in prior papers.[9,12,14] Photo-Hall measurements showed that above 80 K the EPC is due to holes.[19,20] Below 80 K, evidence about *p*-type conduction exists, but recent results by Dischler and Fuchs[21] demonstrated the existence also of free electrons in some LEC samples.

In addition to this the main characteristics of the EPC can be summarized as follows.

(1) It consists of a monotonic increase of the photocurrent.

(2) Once the EPC is reached it remains as long as the temperature is kept below 140 K.

(3) No simultaneous optical-absorption effects have been reported.

(4) Optical quenching of the EPC is achieved with 0.9-eV photons.[10,12]

(5) The EPC is strongly sample dependent, and there exist noticeable differences between HB and LEC samples. HB specimens only exhibit EPC above a certain threshold temperature, which is dependent on sample but lies around 50 or 60 K. The LEC samples normally exhibit the EPC at lower temperature as well. In addition to this they exhibit persistent photocurrent[1,9] (PPC), which has never been observed in the HB samples studied.

This phenomenon shows peculiar characteristics that require special care in interpreting. First of all, some discussion concerning the origin of the excess free carriers is needed, whether they are thermally or optically emitted into the bands.

The correlation between the TSC spectrum as obtained for strong 1.06-μm excitation at 4 K and the I_{ph} versus T plot obtained under saturated illumination with this very wavelength suggests that the EPC is due to a thermal emission of holes (Fig. 4); this is also supported by the absence of optical-absorption-related observations. In spite of this, it should be noted that under short time excitation the hole population seems to be controlled by the optical excitation. This would suggest that the holes are optically transferred in the traps emitting holes. The correlation between the optical absorption and the EPC is an important point of contention.

It should be noted that such a strong hole emission cannot be achieved without previous excitation with 1–1.3-eV light; in other words, it is not possible to fill these acceptors with other light wavelengths, such as 0.8 or 1.45 eV, as long as previous intense excitation with 1–1.3 eV have not been done. This seems to imply that the enhanced photocurrent, though controlled by the occupation of the acceptors, is not merely a trap-filling effect. In other words, it can be argued that the acceptors are activated as a consequence of the metastability. The metastable state can have associated acceptor levels, which are indirectly filled by the optical excitation; the EPC is observed when these acceptors are thermally emptied.

The EPC effect exhibits more complex behavior in LEC samples. The threshold temperature for the observation of the EPC in these samples is significantly lower than it is for the HB specimens, which supports the hypothesis that the shallow levels to be compensated are different from each other. In this sense it is worth noting that HB samples are Cr doped, which provides a high concentration of Cr-related deep acceptor levels that warrant a different compensation level scheme than that of the LEC samples. In addition to this, the LEC samples exhibit persistent photocurrent (PPC) when they are in the EPC state,[1,9] contrary to what happens for the HB samples, for which no persistent photocurrent (PPC) effect was observed.

The PPC effect above 70 K has been tentatively explained by means of the screening of recombination centers by nearby positively ionized shallow donors.[9] It was shown that upon photoneutralization of such donors the PPC level was significantly decreased. This would imply that thermally generated excess holes cannot recombine while the shallow donors are ionized or the temperature is below 130 K, the temperature at which the acceptors are no longer observed (the metastability is thermally destroyed).

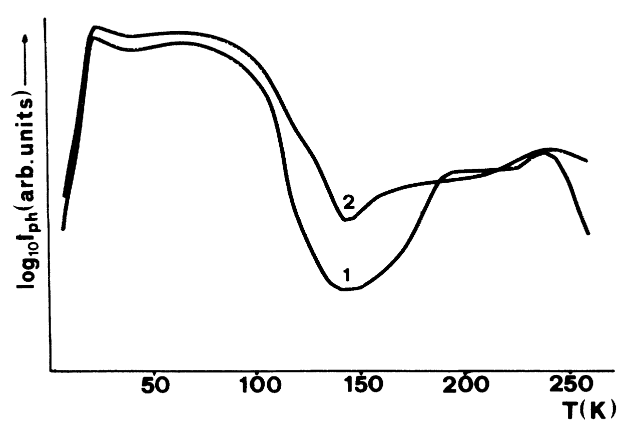

FIG. 4. (1) TSC after YAG (1.06 μm) excitation at 4 K. (2) I_{ph} (1.45 eV) vs t after the previous YAG excitation at 4 K.

Concerning the temperature dependence of the PPC, it must be said that the characteristics of this effect at liquid-helium temperature are different from those reported up until now. Indeed, the photocurrent exhibits a practically nondecaying persistence at 4 K. It should be noted that at this temperature the shallow donors might not be ionized, and the screening would be necessarily less efficient than that reported at 80 K. This is at odds with the markedly nondecaying PPC at low temperature. On the other hand, the excitation with band-gap light does not produce any noticeable decay of the PPC as was the case above 60 K. This could be due to the existence of a repulsive barrier that prevents the capture of free holes at the metastable state, thereby enlarging the lifetime independently of the screening by ionized donors, which will be the dominant mechanism above 60 K, the temperature at which the repulsive barrier of the metastable state itself can be thermally surpassed.

There is another way to quench the low-temperature PPC. Indeed, this can be performed by heating in darkness above 50–60 K. For this purpose the following thermal cycling experiment was carried out.

(1) The sample is illuminated with 1.1-eV light at 4 K until the EPC is attained.

(2) The excitation is removed, and the PPC level is measured.

(3) The sample is heated in darkness up to a temperature T_a and then cooled down to 4 K; the remaining PPC level is measured again.

(4) The sample is heated in darkness above 150 K in order to erase any photomemory effect.

(5) A new cycle is performed, for which the temperature T_a is changed.

This experiment allows us to observe that the PPC is completely quenched around 60 K (Fig. 5). This unambiguously confirms that the low-temperature persistency is not due to the same electronic mechanism displaying the persistency observed above 60 K.

The existence of a persistent carrier population after photoexcitation at 4 K has been reported by other authors, who have also found that such carriers disappear in this temperature range.[6,15] It should be noted that this temperature is well below the thermal restoration threshold of the normal state, which occurs around 130 K, so the fact that the free holes are no longer observed cannot be ascribed to a restoration of the normal configuration but rather to the capture of such holes elsewhere. It is important to remember that the change from the low-temperature ($T < 60$ K) PPC to high-temperature ($T > 60$ K) PPC occurs at the temperature at which the so-called f_0 hole trap emits.[22] The density of this trap has been estimated to be around 10^{16} cm^{-3}, which is high

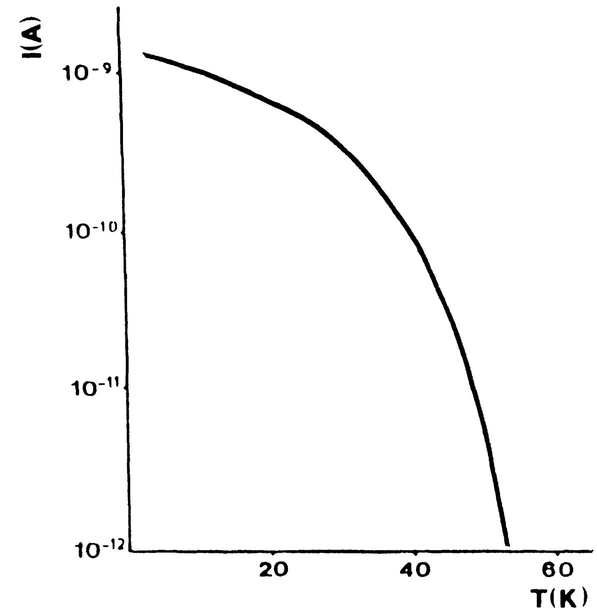

FIG. 5. Persistent photocurrent level at 4 K as a function of the temperature of heating in the dark, labeled T_a in the text.

enough to control the location of the Fermi level. Its activation energy lies around 0.08 eV, which is in agreement with other results and with the $(-/0)$ energy level of the gallium antisite.[23] As has been shown before, this trap is strongly enhanced by the 1–1.3-eV excitation;[4,17] once this has been done it becomes possible to fill the trap by exciting it with light of another wavelength such as 1.45 eV. Without previous 1–1.3-eV light excitation, no matter how long and intense the 1.45-eV excitation was, the concentration of this trap remains several orders of magnitude below that estimated for the EPC situation. All this seems to agree with the observation of intense photogenerated EPR signals, the so-called *FR*1, *FR*2, and *FR*3 centers[24] as well as the intense TSC peaks observed in similar excitation conditions by other authors.[4,17,22,25] A tentative explanation of this is the existence of charge transfers, as suggested before,[12] between the components of the metastable complex, which allows the capture of holes at deep acceptors that then become paramagnetic. In relation to this it is worth noting that the *EL*6 level is characterized by a large difference between its thermal ($\sim$0.35-eV) and optical ($\sim$0.8-eV) activation energies that would imply a large lattice relaxation, which has not been experimentally assessed by uniaxial stress experiments.[26] Nevertheless it has been shown that this level undergoes a multistep electron emission process involving thermally assisted tunneling to nearby shallow levels,[27] which could account for the absence of a strong lattice relaxation, and, on the other hand, evidences the complex nature of the defect. The lattice relaxation could be related to other parts of the defect that are not directly reached by optical measurements as suggested by Levinson.[26] In this interpretation, it should be remembered that the optical quenching of the enhanced photocurrent can be achieved by exciting with 0.9–0.95-eV light,[10,12] which fits the reversible optical quenching of the EPR signal of the (0/+) paramagnetic state of the As_{Ga} defect quite well.[28] This suggests that the optical quenching might be due to an optical transition from the valence band to the (0/+) level of the As_{Ga} defect, seen as a part of the metastable complex displaying the EPC. We will label it as As^*_{Ga} (0/+). This suggests that the defect has a non-EPC configuration, in other words a configuration unable to emit holes. In this observation lies one of the main differences with the excess hole population related to the metastable *EL*2 level. In fact, the spectral distribution of the light restoring the previous configurations is different in each case (Fig. 6). It is clearly seen that the recovering light is not operating at the same electronic level, the (0/+) level of the As_{Ga} for the EPC, but at an unknown level for the metastable *EL*2 level.

Besides this, the optical quenching of the EPC is thermally activated; the hole population associated with EPC cannot be optically quenched below 30 K, and it is almost fully recoverable above 70 K.[10,12] This is in agreement with the thermally activated quenching of the low-temperature persistent free-hole population (Fig. 5). Because the 0.9-eV excitation excites electrons to the (0/+) level of the As^*_{Ga} defect, and this is not observed at low temperature, it is reasonable to assume that only when the As^*_{Ga} is positively ionized is it possible optically to quench the hole population, and this could be done by the thermally activated capture of low-temperature ($T < 60$ K) holes at the metastable centers. Such a process can be described as follows: both below and above 70 K holes are transferred to the acceptor levels by exciting with 1–1.3 eV light the metastable state. The emitted holes are captured at the metastable levels, so the EPC would disappear. Below 60 K, this capture is less probable because of the barrier associated with the metastable state itself, as the temperature is decreased the per-

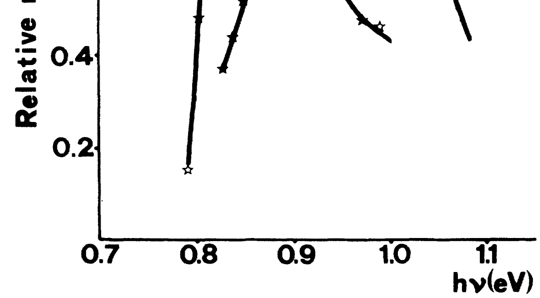

FIG. 6. Spectral distribution for the recovery of the normal configurations of *EL*2 (PQ) (open stars) and *EL*6 (EPC) (solid stars).

sistence (PPC) is more evident. On the other hand, the lowering of the hole capture efficiency at the metastable state, as well as the lower acceptor-hole emission efficiency at low temperature seems to suggest that the hole population on the metastable state, and therefore the ionized As_{Ga}^{*+} concentration, is low enough, which should justify the low recovery efficiency of the state before EPC by 0.95-eV excitation, which is unobservable below 30 K and considerably reduced between 30 and 60 K. Above 60 K the holes are thermally emitted from the acceptor traps and captured at neutral As_{Ga}^{*}. Then the efficiency of the recovery with the 0.95-eV light increases. The EPC is associated with the thermal emission of such holes. The 1–1.3-eV excitation restores the hole population trapped at the acceptors.

It should be noted that the semi-insulating HB material is Cr doped, and that probably the deep acceptor levels introduced by Cr capture the free holes responsible for the EPC, thus avoiding the existence of a persistent hole population. On the other hand, the existence of LEC samples that only exhibit the photoquenching effect, the EPC effect being quite weak in these samples, rules out the possibility that the excess hole population providing the EPC would be the consequence of the metastability of *EL*2. Indeed, if the existence of such holes were the only consequence of the lack of compensation of the acceptors when the *EL*2 level is transformed into the metastable state, they would also be observed in these samples because the compensation ratio $[EL2]+[N_D]/[N_A]$ must be preserved and the compensated acceptors must always exist. In spite of this, an important point of contention is related to the role played by the metastability of the *EL*2 level in the EPC even if both effects seem to be associated with different relaxations. An important question to be answered is this: Could the EPC be observed without a previous *EL*2 level photoquenching? In other words, are the changes in compensation introduced by the metastability of the *EL*2 level necessary for observation of the EPC? Investigations around this point are now in progress.

IV. CONCLUSION

The members of the excess hole population induced by light excitation with 1–1.35-eV light have different origins depending on the samples and the experimental conditions of the optical excitation producing it. The holes directly related to the photoquenching are due to uncompensated acceptors, and a part of the expected hole population is trapped at deeper acceptor levels, which seems to be associated with the metastability of *EL*2.

The excess hole population related to the EPC is mainly due to the thermal emission of holes from traps ($E_t < 120$ meV), which are related to an optically induced metastable state, probably associated with *EL*6. The mechanisms displaying the persistent photocurrent (PPC) are different below and above 60 K.

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