Piezoelectric photoacoustic studies of optical recovery of metastable states related to EL2 and EL6 levels in semi-insulating GaAs

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Effect of secondary-light illuminations of $h\nu=0.9$ and 1.45 eV on photoquenched and photoenhanced states in semi-insulating GaAs are investigated using piezoelectric photoacoustic (PPA) measurements at 80 K. It is found that the secondary-light illuminations of 0.9 and 1.45 eV cause an optical recovery of the PPA signal from EL2* to $EL2^0$. We observed an appearance of a broad band at 0.8–1.0 eV in the PPA spectrum after the secondary-light illumination of 0.9 eV. The most important finding is that the PPA spectra after the secondary-light illumination of 0.9 eV both on the photoquenched and on the photoenhanced states are the same in the whole photon-energy region. We conclude that the secondary-light of $h\nu=0.9$ eV induces both an optical recovery and a generation of metastable state of the EL6 level at the same time. The different rates for the transformation of these two processes explain well the observed complex natures of the PPA signal under the secondary-light illumination. [S0163-1829(98)04343-4]

I. INTRODUCTION

Piezoelectric photoacoustic (PPA) spectroscopy has recently been used for investigating physical properties of semiconductors. One of the great advantages of the PPA spectroscopy is that it is a direct monitor of the nonradiative recombination processes. The other advantage is that the PPA spectroscopy is sensitive to a very small optical absorption coefficient in a highly transparent sample. The presence of these two great advantages indicates that PPA spectroscopy would be a very useful tool for investigating deep levels in GaAs. In our previous papers, we reported that the electron transition involving EL2 in a semi-insulating (SI) GaAs sample could be observed by using PPA measurements.[1,2] The usefulness of the PPA technique for the characterization of the deep levels were also demonstrated.

Deep-lying defect level EL2 is known to be a dominant donor to accomplish a SI nature of GaAs substrate for large scale integration applications. Since EL2 transforms to its metastable state by the monochromatic-light illumination about 1.1 μm at the low-temperature, so-called photoquenching effect,[3] its electronic structure has been extensively studied. One of the commonly accepted properties about this metastable state (EL2*) is that the initial state before photoquenching ($EL2^0$) can be revived after annealing the sample above 130 K. In addition, the existence of an optical recovery from EL2* to $EL2^0$ was observed in SI samples by Manasreh and Fischer.[4] Although they reported that photons around 0.9 eV and/or ranging from 1.4 to 1.51 eV enable us to resume $EL2^0$ partly, to our knowledge, no thorough discussions were carried out yet.

Jiménez, Alvárez, and Bonnafé[5] reported a very strong enhancement of the photocurrent (EPC) by the prolonged monochromatic-light illumination about 1.1 μm after photoquenching of EL2. They concluded that EPC was attributed to the EL6 level that is ubiquitous in bulk GaAs. They also reported an additional optical quenching of EPC by the secondary-light illumination of $h\nu=0.9$ eV.[6] However, to our knowledge, EPC and its optical quenching in the photoconductivity measurements have never been confirmed by other experiments.

We have already reported that there are two kinds of photoinduced states of the PPA signal in SI GaAs after the quenching-light illumination of $h\nu=1.12$ eV at 80 K.[7] One is the quenched state generated by a short period illumination. The other is the enhanced state attained by a prolonged illumination. Since we will report on the effect of the secondary-light illumination on these two photoinduced states, we briefly review the previous results to make clear the current discussions. In this paper, we refer to these two photoinduced states as Q and E states, respectively. Both states were effectively generated when the quenching light was set in the similar photon-energy region from 1 to 1.2 eV. The typical PPA spectra of the state before the quenching-light illumination, referral to as the N state, and those of the two photoinduced states are shown in Fig. 1. A hump above 1.0 eV up to the band-gap energy ($E_g=1.51$ eV at 77 K) is observed in the PPA spectrum of the N state as shown by the solid curve. We concluded in our previous papers that this hump was caused by the electron nonradiative transitions involving the EL2 level.[1,2] Vanishing of this hump occurs by the quenching-light illumination for a short period of 2 or 3 min because of the photoquenching effect of EL2 (Q state, shown by the broken curve). The E state produced by the quenching-light illumination for a long time shows a wide band over the range from 0.6 to 1.4 eV as shown by the dotted curve in the figure. We refer to this wide band as the E band.

A typical time dependence of the PPA signal during the quenching-light illumination is shown in Fig. 2. After the sample was cooled down to 80 K in the dark, the quenching light illuminated on sample. The PPA signal intensity at $h\nu=1.12$ eV was recorded as a function of illumination time.

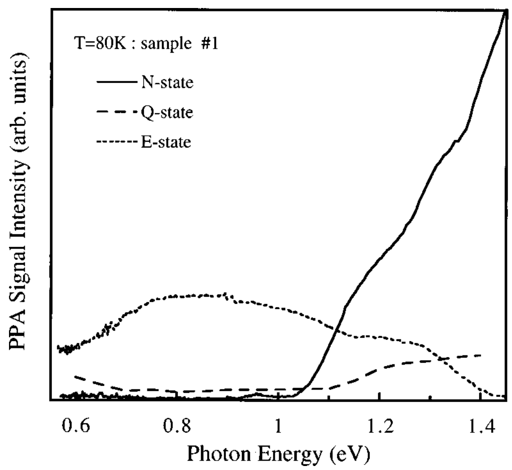

FIG. 1. The typical PPA spectra of the *N*, *Q*, and *E* states at 80 K.

As shown in Fig. 2, the time-dependent PPA signal shows the complex feature specified by three states as labeled in the figure. The PPA signal intensity at this photon-energy region in GaAs is considered to be proportional to the absorption coefficient and to the total EL2 concentration.[7] A decrease of the PPA signal due to photoquenching of EL2 is observed for a short period of 2.5 min. This state corresponds to the *Q* state in Fig. 1. Strong enhancement of the PPA signal after fully photoquenching is observed by the prolonged quenching-light illumination. If the temperature of the sample was kept at 80 K both photoinduced states were quite stable. The *N* state could be obtained by heating the sample in the quenched and/or enhanced states above 130 K and subsequent cooling down to 80 K. This means that a thermal recovery from the *Q* and/or *E* states to the *N* state occurs around this temperature.

The presence of the deep donor EL6 level and its metastable state $EL6^m$ were proposed to explain the *E* state of the PPA signal. EL6 donates electrons to compensate a part of the carbon acceptor after the $EL2^0$ to $EL2^*$ transformation is accomplished. In this situation, positively ionized $EL6^+$ can transform to metastable $EL6^m$, which becomes optically active by the quenching-light illumination for a long time. The nonradiative recombinations of photoexcited electrons from the valence band (VB) and/or from the compensated carbon acceptor to $EL6^m$ take place. These nonradiative recombinations are detected as the *E* band in the PPA spectrum of the *E* state.

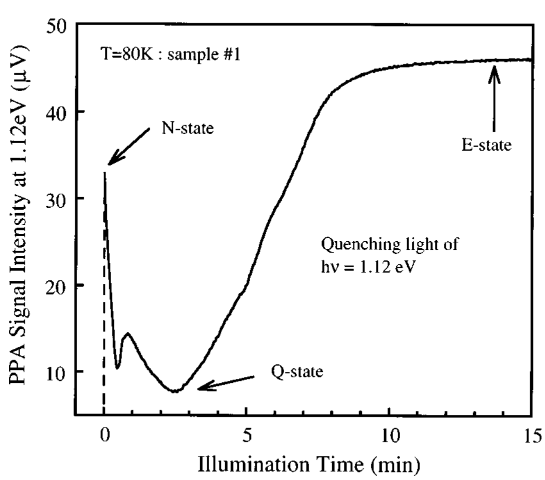

FIG. 2. The typical time-dependent PPA signal at 1.12 eV during the quenching-light illumination of $h\nu = 1.12$ eV. The PPA signal shows the complex feature specified by three states as labeled in the figure.

In this paper, we report the effects of the secondary-light illuminations of $h\nu = 0.9$ and 1.45 eV on the two photoinduced states observed in the PPA spectra at low temperature from a nonradiative recombination point of view. Optical recovery from $EL2^*$ to $EL2^0$ was observed by illuminating the secondary light with photon energy of $h\nu = 0.9$ and/or 1.45 eV. In addition, we also observed an appearance of a broad band at 0.8–1.0 eV in the PPA spectrum after the secondary-light illumination of 0.9 eV on both photoinduced states. The most important finding was that the PPA spectra after the secondary-light illumination of $h\nu = 0.9$ eV on both photoinduced states were the same in the whole photon-energy region. We concluded that the secondary light of $h\nu = 0.9$ eV induced both an optical recovery and generation of metastable state of the EL6 level. The different rates of the transformations for these two processes explain well the observed complex natures of the PPA signal under the secondary-light illumination.

II. EXPERIMENTAL PROCEDURES AND RESULTS

Two samples were prepared from carbon-concentration controlled SI GaAs wafers grown by a liquid-encapsulated Czochralski (LEC) method. The carbon concentrations in samples 1 and 2 were 1.1 and $0.4 \times 10^{16}\ \mathrm{cm}^{-3}$, respectively. The total EL2 concentrations in the two samples were the same at $(1.2-1.4) \times 10^{16}\ \mathrm{cm}^{-3}$. Since these wafers were thermally treated by a three-stage annealing method,[8] a minimum amount of irrelevant intrinsic defects was expected. Details of the experimental setups for the PPA measurements are reported elsewhere.[9] The quenching and the secondary light were provided from a monochromator with an external 150-W halogen lamp. The photon energy of the quenching light was set at 1.12 eV, while that of the secondary light was set at 0.9 or 1.45 eV. The illuminations were carried out at 80 K. We hereafter refer to the step of the secondary-light illumination for 30 min at 80 K as the SLI.

A sequence of measuring an effect of the SLI of $h\nu = 0.9$ eV on the *Q* state is shown in Fig. 3. After the sample was cooled down to 80 K in the dark, the quenching light of 1.12 eV was illuminated on the sample for 2.5 min to transform the sample to the *Q* state and was then cut off (curve 1 in Fig. 3). Next, SLI of $h\nu = 0.9$ eV was carried out with keeping the temperature of the sample at 80 K (curve 2 in Fig. 3). At this time, both the probe and the secondary light were set at the photon energy of 0.9 eV. Finally, the time-dependent PPA signal intensity at 1.12 eV was measured again with illuminating the quenching light. The result is shown by curve 3 in Fig. 3. The PPA signal intensity has a value of that in the *N* state first and exhibits the photoquenching and following enhancement as in the case of Fig. 2. This experimental result implies that the secondary light with photon energy of 0.9 eV causes an optical recovery from the *Q* state to the *N* state.

In the same way as the *Q* state, the effect of the SLI of 0.9

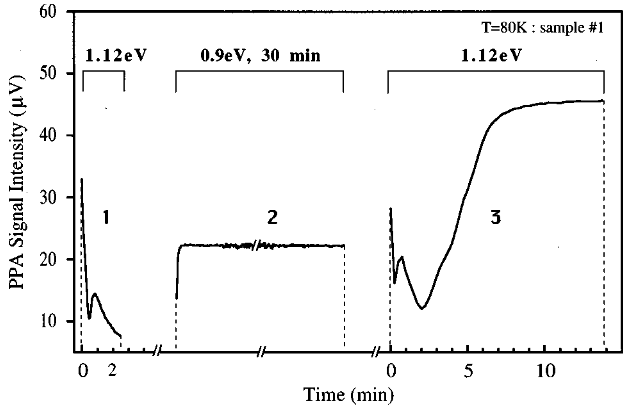

FIG. 3. A sequence of measuring the effect of the SLI of 0.9 eV for the *Q* state at 80 K. After the sample was cooled down to 80 K in the dark, the quenching light of 1.12 eV was illuminated for 2.5 min to transform the sample to the *Q* state (curve 1). Next, the secondary light of $h\nu = 0.9$ eV was illuminated for 30 min with keeping the temperature at 80 K (curve 2). Finally, the time-dependent PPA signal at 1.12 eV was measured again with illuminating the quenching light (curve 3).

eV on the *E* state was measured. A sequence is shown in Fig. 4. The quenching light was first illuminated on the sample for 15 min to transform to the *E* state (curve 1 in Fig. 4). The secondary light with photon energy of $h\nu = 0.9$ eV was illuminated on the sample for 30 min (curve 2 in Fig. 4). The time dependence of the PPA signal intensity at 1.12 eV was then measured as a function of the quenching-light illumination time and the result is shown by curve 3 in Fig. 4. Recovery of the PPA signal to the *N* state at 1.12 eV and following photoquenching and enhancement of the PPA signal are observed. This is the same as that in the case on the *Q* state. This implies that an optical recovery from the *E* state to the *N* state occurs by the SLI of 0.9 eV.

The time dependences of the PPA signal under the quenching-light illumination from the *N* state (curve 1 in Fig. 3) and optically recovered states from the *Q* state (curve 3 in Fig. 3) and from the *E* state (curve 3 in Fig. 4) by the SLI of 0.9 eV are replotted in Fig. 5 to compare with each other. For the optically recovered states, photoquenching and enhancement by the quenching-light illumination occur faster than that for the *N* state. It is also found that signal intensities at the *Q* state in two curves after the SLI of 0.9 eV (broken and dotted curves) are larger than that in the *N* state (solid curve), while the signal intensities at the *E* state in three curves are the same.

The effect of the SLI of $h\nu = 0.9$ eV on the PPA spectrum of the *Q* state was also measured. The sample was first cooled down to 80 K in the dark and underwent to the *Q* state by the quenching-light illumination for 2.5 min. Next, the SLI of $h\nu = 0.9$ eV was carried out with keeping the temperature at 80 K. Finally, the PPA spectrum was measured as a function of photon energy of the probe light ranging from 0.6 to 1.45 eV. It was also confirmed that there was no effect of the probe light during the spectrum measurements. The probe light did not cause any further transformation on the states. The result is shown in Fig. 6 by the open circles. As shown in the figure, a broad band at 0.8–1.0 eV and a hump

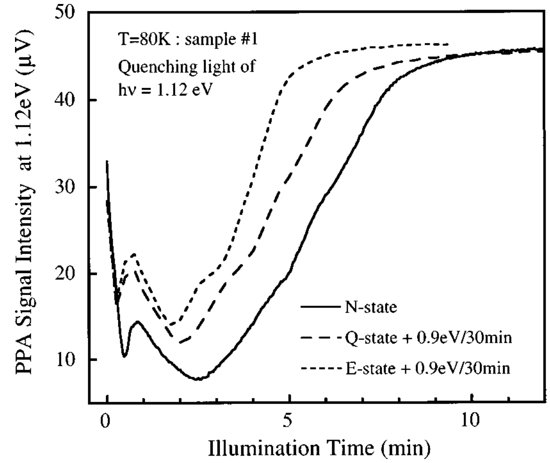

FIG. 5. The time dependences of the PPA signal under the quenching-light illumination from the *N* state (curve 1 in Fig. 3), the *Q* state after the SLI of 0.9 eV (curve 3 in Fig. 3), and the *E* state after the SLI of 0.9 eV (curve 3 in Fig. 4) are replotted again in Fig. 5.

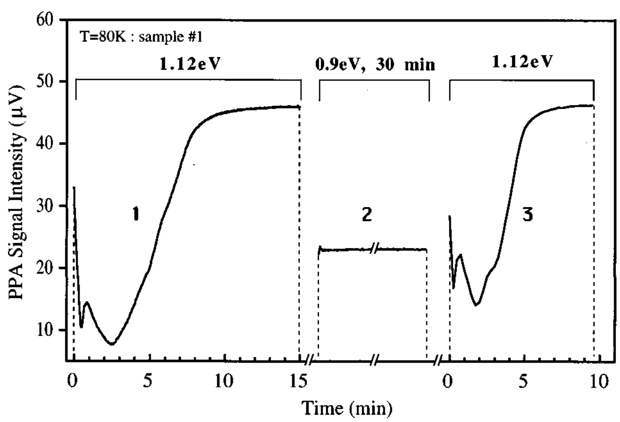

FIG. 4. A sequence of measuring the effect of the SLI of 0.9 eV for the *E* state at 80 K. The same manner was used as in the case for the *Q* state. The quenching light of 1.12 eV was illuminated for 15 min to transform the sample to the *E* state (curve 1).

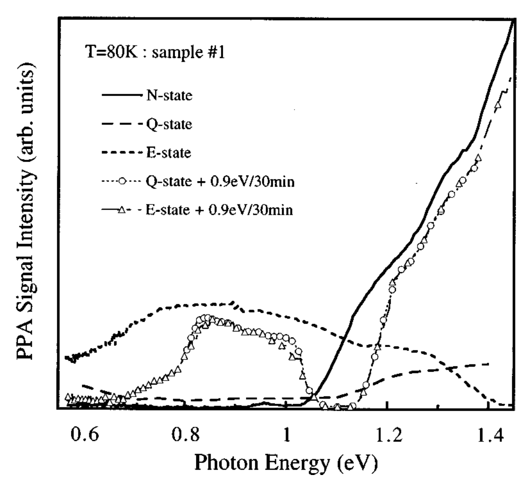

FIG. 6. The PPA spectra after the secondary-light illumination of 0.9 eV on the *Q* and *E* states for 30 min. A broad band at 0.8–1.0 eV and a hump above 1.1 eV up to the band-gap energy are observed. The PPA spectra of the *N*, *Q*, and *E* states at 80 K are also shown.

above 1.1 eV up to the band-gap energy appear. The PPA spectra of the *N* and *Q* states are also shown in the figure for the sake of comparison. It seems that the PPA spectrum in the higher photon-energy region above 1.1 eV almost recover to that of the *N* state by the SLI of 0.9 eV on the *Q* state. However, the broad band at 0.8–1.0 eV appears in the lower photon-energy region. We hereafter refer to this broad band as the *A* band. It was found that after the sample was annealed at 130 K for a few min and was subsequently cooled down to 80 K, the *A* band completely vanished and the PPA spectrum of the *N* state was obtained again.

The PPA spectrum after the SLI of 0.9 eV on the *E* state for 30 min was also measured in the same manner with the case for the *Q* state. The result is shown in Fig. 6 by the open triangles. The PPA spectrum of the *E* state is also shown in the figure by the dotted curve. In Fig. 6, a hump above 1.1 eV up to the band-gap energy appears in the higher photon-energy region. An optical recovery from the *E* state to the *N* state may occur partly in the higher photon-energy region above 1.1 eV by the SLI of 0.9 eV. A broad band at 0.8–1.0 eV is still observed in the lower photon-energy region and the intensity and the spectral width are small compared with that of the *E* band in the PPA spectrum of the *E* state. Although the spectral shape is similar to that of the *A* band, we label this broad band at 0.8–1.0 eV as the *B* band. It was found that after the sample was annealed at 130 K for a few min and was subsequently cooled down to 80 K, the *B* band completely vanished and the PPA spectrum showed the *N* state. It is found from Fig. 6 that the PPA spectra after the SLI of 0.9 eV on the *Q* and *E* states are the same in the whole photon-energy region.

The effect of the SLI was also measured when the secondary light is set at the photon energy of 1.45 eV. The experimental procedures were the same as that when the secondary light was set at 0.9 eV. The sample was first cooled down to 80 K in the dark and underwent to the *Q* or *E* state by the quenching-light illumination for respective time. Next, the SLI of 1.45 eV on the *Q* or *E* states was carried out with keeping the temperature at 80 K. After that, the time-dependent PPA signal intensities at 1.12 eV and the PPA spectra as a function of photon energy of the probe light ranging from 0.6 to 1.45 eV were measured. The time-dependent PPA signal after the SLI of 1.45 eV on the *Q* and *E* states are shown in Fig. 7 by the open circles and triangles, respectively. The PPA signal after the SLI of 1.45 eV on the *Q* state starts from the midvalue smaller than that of the *N* state. This means that the *Q* state is not completely recovered by illuminating the secondary light of 1.45 eV. On the contrary, no effect of the SLI of 1.45 eV on the *E* state can be observed. The PPA signal intensity remains constant even after the SLI of 1.45 eV.

The effects of the SLI of 1.45 eV on the PPA spectra of the *Q* and *E* states are shown in Fig. 8. The PPA spectrum above 1.1 eV is slightly increased by the SLI of 1.45 eV on the *Q* state for 30 min as shown by the open circles. The PPA spectra of the *N* and *Q* states are shown by the solid and broken curves, respectively. This result is in agreement with the result in the time-dependent PPA measurement in Fig. 7. The value at $t=0$ after the SLI of 1.45 eV on the *Q* state is slightly larger than the value of the *Q* state. On the contrary, the PPA spectrum of the *E* state is not influenced by the SLI

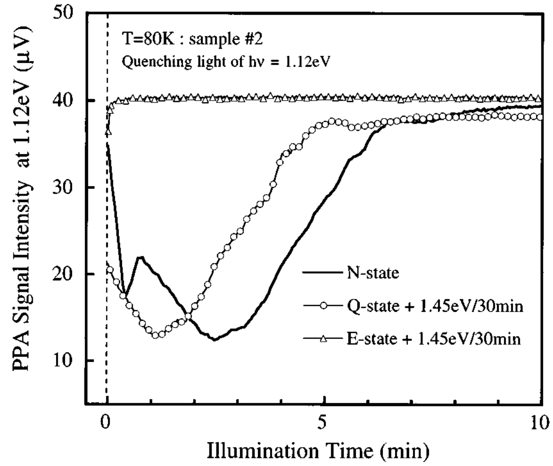

FIG. 7. The time-dependent PPA signal of 1.12 eV after the secondary-light illumination of 1.45 eV on the *Q* and *E* states for 30 min. The PPA signal intensity increases from a value of the *Q* state. On the other hand, the secondary-light illumination of 1.45 eV does not affect the *E* state. The PPA signal intensity shows a value of the *E* state even after the secondary-light illumination for 30 min as shown by the open triangles.

of 1.45 eV for 30 min. The spectrum is the same as that of the *E* state as shown by the open triangles. This is also consistent with the result of the time-dependent PPA measurements shown in Fig. 7.

III. DISCUSSIONS

We could observe the effects of the SLI of 0.9 eV or 1.45 eV on two photoinduced states by measuring the time-dependent and spectral PPA signals. As we have reported in our previous paper,[7] two photoinduced states are due to metastable transformations of EL2 and EL6, respectively. To clarify the effects of the SLI on these defect levels in more detail, we will first discuss the SLI on the *Q* state.

A. Secondary-light illuminations on the *Q* state

From the time-dependent PPA measurement, as shown in Fig. 3, a recovery of the PPA signal intensity at 1.12 eV to a

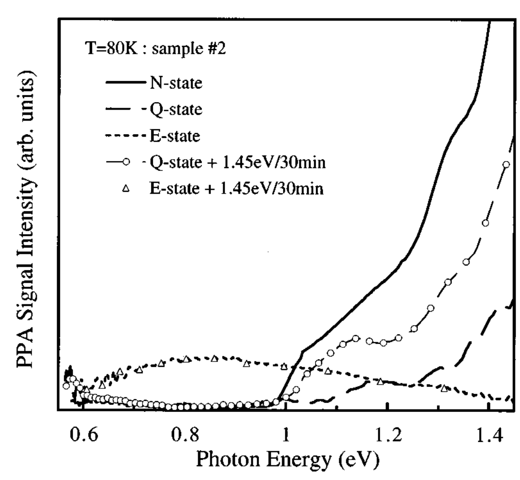

FIG. 8. The PPA spectrum after the secondary-light illumination of 1.45 eV on the *Q* state for 30 min. An optical recovery of the PPA signals are observed.

value the of the *N* state is observed by the SLI of 0.9 eV on the *Q* state. In Fig. 6, the PPA spectrum is also recovered to that of the *N* state after the SLI of 0.9 eV on the *Q* state. These experimental results imply that the secondary light with a photon energy of 0.9 eV causes an optical recovery from the *Q* state to the *N* state. We have already reported that the PPA signals below the band-gap energy region in the *N* state are caused by the nonradiative recombinations of electrons involving the EL2 level, and that the *Q* state is the result of the photoquenching of EL2.[1,2] Therefore, the increase of the PPA signals above 1.1 eV implies that an optical recovery from EL2* to $EL2^0$ occurs by the SLI of 0.9 eV on the *Q* state at 80 K.

When the secondary light was set at the photon energy of 1.45 eV, an optical recovery also occurred. In Fig. 7, the PPA signal after the SLI of 1.45 eV on the *Q* state increases to about 20 μV. When the quenching light is further illuminated, the PPA signal begins to decrease due to the photoquenching of EL2. This partial recovery of the PPA spectrum from the *Q* state is also shown in Fig. 8 by the open circles. The PPA spectrum has almost the same shape above 1.1 eV as that of the *N* state. A complete recovery of the *N* state was accomplished by heating the sample at 130 K for a few min and was subsequently cooled down to 80 K. Although the effectiveness of the optical recovery is smaller than in the case of 0.9 eV, we consider that the optical recovery from the *Q* state occur certainly when the photon energy of the secondary light is set at 1.45 eV.

Manasreh and Fischer reported by using infrared optical absorption measurements that a partial optical recovery from EL2* to $EL2^0$ occurred by the monochromatic-light illumination of $h\nu = 0.9$ eV and/or ranging from 1.4 to 1.51 eV.[4] It is noted here that the observed features of the optical recovery in our PPA measurements are similar to those observed in the infrared optical-absorption measurements except for the appearance of the *A* band. Although the signal-generation mechanisms are different among the two experiments, it can be considered that the effects of the SLI on the photoquenched state may be the same. Therefore, we conclude that the secondary-light illumination of 0.9 and/or 1.45 eV on the *Q* state for 30 min at 80 K causes an optical recovery from EL2* to $EL2^0$. It is found from the present experimental results that the light with photon energy of 0.9 eV is more effective than in the case of 1.45 eV. To our knowledge, this is the first time that an optical recovery of EL2 from its metastable state is observed from a nonradiative recombination point of view.

It is noted that the *A* band appeared in the PPA spectrum after the SLI of 0.9 eV is not observed in the PPA spectrum of the *N* or *Q* state. In addition, when the secondary light is set at 1.45 eV, the *A* band does not appear. If the sample was heated to 130 K and subsequently cooled down to 80 K, the *A* band vanished and the sample showed the PPA spectrum of the *N* state. These experimental results cannot be explained by simply supposing that the SLI of 0.9 eV causes an optical recovery of EL2 as discussed above. It seems that the SLI of 0.9 eV on the *Q* state produces a subsidiary effect. Since the similar broad band is also observed after the SLI of 0.9 eV on the *E* state, we will discuss below about the two broad bands in more detail.

B. Secondary-light illumination on the *E* state

We next discuss the effects of the SLI of the *E* state. A recovery of the 1.12 eV PPA signal intensity to a value of the *N* state is observed by the SLI of 0.9 eV on the *E* state as shown by curve 3 in Fig. 4. After the recovery, the PPA signal at 1.12 eV shows the typical time dependence of the photoquenching and following enhancement by the continuous quenching-light illumination. This experimental result suggests that an optical recovery to the *N* state also occurs even from the *E* state by the SLI of 0.9 eV. Indeed, the PPA spectrum after the SLI of 0.9 eV on the *E* state shows the optical recovery to that of the *N* state in the higher photon-energy region above 1.1 eV as shown in Fig. 6. We have already reported in our previous paper[7] that when the sample is in the *E* state, two metastable states of EL2* and $EL6^m$ exist. Therefore, it is necessary to consider the effects of the SLI on both EL2* and $EL6^m$ at the same time. As discussed above, EL2* can optically recover to $EL2^0$ by the SLI of 0.9 eV or 1.45 eV on the *Q* state. Since the PPA signal after the SLI of 0.9 eV on the *E* state shows the same time dependence as that of the *N* state (shown by curve 3 in Fig. 4), an optical recovery to the *N* state should occur. Therefore, we can consider that optical recoveries both from EL2* to $EL2^0$ and from $EL6^m$ to $EL6^+$ occur at the same time by the SLI of 0.9 eV on the *E* state. In the lower photon-energy region below 1.1 eV, the *B* band appears after the SLI of 0.9 eV on the *E* state. This band cannot be observed in the PPA spectrum of the *N* state. If the optical recovery of both EL2 and EL6 are completely accomplished, the spectrum should not show the *B* band. Since the thermal recovery above 130 K always change the spectrum to that of the *N* state, the present optical recovery to the *N* state may not be completed.

On the other hand, when the secondary light is set at the photon energy of 1.45 eV, the PPA signal in the *E* state remains unchanged by the SLI as shown in Fig. 7 (the open triangles). If the sample was annealed at 130 K for a few min and was subsequently cooled down to 80 K, the PPA signal showed the same time dependence as that of the *N* state. The PPA spectra before and after the SLI of 1.45 eV on the *E* state are also the same as shown in Fig. 8. These imply that metastable EL2* and $EL6^m$ still exist even after the SLI of 1.45 eV. This is quite different from the case for the *Q* state, where an optical recovery occurs both for the secondary light at 0.9 and 1.45 eV.

In the present experimental results, the most important finding is that both the PPA spectra after the SLI of 0.9 eV on the *Q* and *E* states for 30 min at 80 K are the same in the whole photon-energy region. The broad band at 0.8–1.0 eV, the *A* or *B* band, and a hump above 1.1 eV up to the band-gap energy appear. Since the thermal stabilities of these state are the same, it can be considered that the states generated by the SLI of 0.9 eV on the *Q* and *E* states are identical. Although the intensity and the spectral width of the *A* and *B* bands are small compared with that of the *E* band in the PPA spectrum of the *E* state, the origin of these broad bands and the *E* band seems to be the same. In order to check this assumption, we carried out an additional measurement. To investigate a change of the PPA spectrum as a function of the time of the SLI on the *E* state, the PPA spectra were measured one by one by illuminating the secondary light for every 30 s. It was found that the overall PPA signals for the

E band decreased monotonously except for the photon-energy region for the *B* band. Therefore, we considered that the appearance of the *B* band was the result of the optical recovery of the *E* band by the SLI of 0.9 eV on the *E* state. In the higher photon-energy region above 1.1 eV, the PPA spectrum first decreased to the spectrum of the *Q* state and subsequently increased to that of the *N* state. This is consistent with the above discussion that in the higher photon-energy region above 1.1 eV, the optical recovery to the *N* state occur even from the *E* state by the SLI of 0.9 eV. The PPA spectrum was finally coincident with that shown in Fig. 6 by the open triangles. Even if the secondary light of 0.9 eV further illuminated on this state more than 30 min, no change in the PPA spectrum was observed.

A change of the PPA spectrum as a function of the time of the SLI of 0.9 eV on the *Q* state was also measured in the same manner for the *E* state. The PPA spectrum above 1.1 eV monotonously increased from the PPA spectrum of the *Q* state with increasing the illumination time. At the same time, an increase of the PPA spectrum at the photon-energy region for the *A* band was observed. As a result, the PPA spectrum after SLI of 0.9 eV on the *Q* state for 30 min coincided with that of the recovered *E* state in the whole photon-energy region as shown in Fig. 6. If the quenching light of 1.12 eV further illuminated on these recovered states, the *A* and *B* bands monotonously increased to form the *E* band in the PPA spectrum of the *E* state, while the PPA spectra above 1.1 eV first decreased to that of the *Q* state and subsequently increased to that of the *E* state. Finally, the PPA spectra showed the *E* state.

From these additional measurements, we conclude that the states generated by the SLI of 0.9 eV on the *Q* and *E* states are identical, and that the optical recovery from EL2* to $EL2^0$ occurs by the SLI of 0.9 eV on both the *Q* and *E* states. The optical recovery increases a number of electrons excited from $EL2^0$ to the conduction band (CB). This results in an increase of the PPA spectrum in the higher photon-energy region above 1.1 eV as shown in Fig. 6. In the lower photon-energy region below 1.1 eV it was also found that the *A* and *B* bands were the intermediate state in the transformation process to the *E* band. Since the *B* band is the result of the partial optical recovery of the *E* band by the SLI of 0.9 eV on the *E* state, we can consider that the origins of two broad bands and the *E* band are the same. In our previous paper,[7] we have reported that the electron transition from VB and/or from the compensated carbon acceptor to $EL6^m$ causes the *E* band. The *A* and *B* bands are also caused by the electron nonradiative transitions involving $EL6^m$. We conclude that the SLI of 0.9 eV on the *E* state induces the optical recovery of $EL6^m$ partly. Since this results in the decrease of electrons excited from VB and/or compensated carbon acceptor to $EL6^m$, the intensity and width of the *E* band decrease except for the photon-energy region for the *A* and *B* peaks.

According to our present model, $EL6^m$ should be generated by the SLI of 0.9 eV on the *Q* state. In our previous paper,[7] we reported that when the sample is in the *Q* state, $EL6^m$ does not exist and that the metastable transformation from $EL6^+$ to $EL6^m$ effectively occurred in the photon-energy region from 1 to 1.3 eV. To explain the present experimental results, we propose here that the SLI of 0.9 eV can induce both the optical generation and dissociation of $EL6^m$. If the sample is in the *E* state, all of EL6 already transforms to $EL6^m$. Therefore, the *E* band partly decreases by the SLI of 0.9 eV due to optical recovery from $EL6^m$ to $EL6^+$ except for the photon-energy region for the *B* band. When the sample is in the *Q* state, all of EL6 exist as $EL6^+$ due to the compensation of the carbon acceptor.[7] A metastable transformation from $EL6^+$ to $EL6^m$ occurs by the SLI of 0.9 eV and results in an increase of the PPA signal in the photon-energy region for the *A* band. The difference rates of the transformation for these two processes of the generation and dissociation of $EL6^m$ explain the observed complex natures of the PPA spectra after the SLI of 0.9 eV. Since the SLI of 0.9 eV also induce an optical recovery of $EL2^0$, the PPA spectra in the higher photon-energy region above 1.1 eV increase at the same time. As a result, the PPA spectra after the SLI of 0.9 eV on the *Q* and *E* states are coincident with each other in the whole photon-energy region as shown in Fig. 6.

It should be noted that the current model is based on one assumption that the presence of metastable EL2* is not a necessary condition for the transformation from $EL6^+$ to $EL6^m$. There is no correlation between the mechanisms of the metastable generation of EL2* and $EL6^m$. However, we have reported in our previous paper that the *E* state could not be achieved without undergoing the *Q* state.[7] In order to check the validity of the former assumption, we investigated an effect of the SLI of 0.9 eV on the *N* state. When the sample is in the *N* state, EL2 and EL6 exist as normal $EL2^0$ and $EL6^+$, respectively. If the metastable transformation from $EL6^+$ to $EL6^m$ does not need a photoquenching situation of EL2, the SLI of 0.9 eV should increase the PPA signal in the photon-energy region for the *A* and *B* bands. After the sample was cooled to 80 K in the dark, the secondary light of 0.9 eV was immediately illuminated on the sample for 30 min. The PPA spectrum was measured as a function of photon energy of the probe light ranging from 0.6 to 1.4 eV. The result is shown in Fig. 9 by the open circles. The PPA spectra after the SLI of 0.9 eV on the *Q* and *E* states and that of the *N* state are shown in the figure to compare the result. A small but certain broad band around

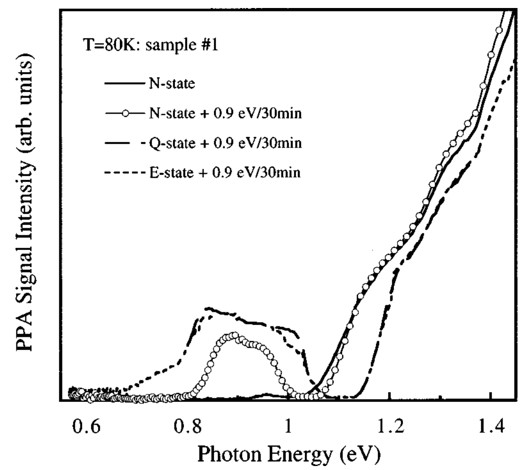

FIG. 9. The PPA spectrum after the secondary-light illumination of 0.9 eV on the *N* state for 30 min. A small but certain broad band around 0.8–1.0 eV appears.

0.8–1.0 eV appears. This broad band was quite stable unless the temperature of the sample increased above 130 K as well as the thermal behaviors of the *A* and *B* bands. It is noted that the PPA signals in the higher photon-energy region above 1.1 eV are not influenced by the SLI of 0.9 eV.

The time-dependent PPA signal after the SLI of 0.9 eV on the *N* state was also measured by using the same manner for the *Q* and *E* states. The PPA signal showed the typical time dependence as for the *N* state. This result is in agreement with the PPA spectrum of the *N* state after the SLI of 0.9 eV. These experimental results suggest that the SLI of 0.9 eV cannot change the situation of the EL2 level. This is consistent with our previous papers that the photoquenching of EL2 occur by illuminating the monochromatic light in the photon energy ranging from 1.0 to 1.3 eV.[3,7] Therefore, we conclude that the metastable transformation from $EL6^+$ to $EL6^m$ can occur even from the *N* state by the SLI of 0.9 eV and that there is no correlation between the metastable transformation mechanisms of $EL6^m$ and $EL2^*$. No matter what the EL2 level is in any situation of normal or metastable, the SLI of 0.9 eV causes the metastable transformation from $EL6^+$ to $EL6^m$ and the PPA signal around 0.9 eV increases.

In Fig. 5, it takes only 2 and 10 min to reach the *Q* and *E* states for the recovered states after the SLI of 0.9 eV on the *Q* and *E* states, respectively. They are faster than that for the *N* state, which is shown in Fig. 2. It is also found that signal intensities at the *Q* state in the open circles and triangles are larger than that in the solid curves, while those at the *E* state are the same. A complex feature of the time-dependent PPA signal at 1.12 eV is due to overlapping of two photoinduced processes.[7] One is the photoquenching of EL2 and the other is the metastable generation of $EL6^m$. When the sample is in the *N* state, EL2 and EL6 exist as normal $EL2^0$ and $EL6^+$, respectively. If the secondary light of 1.12 eV illuminates, the metastable transformations both for EL2 and EL6 occur. The delay of the metastable transformation of $EL6^m$ than that of EL2 can be explained by assuming that the metastable transformation probability of EL2 is larger than that of EL6. After the SLI of 0.9 eV on the *Q* and *E* states, the situation changes. According to the above discussions, a part of EL6 already exists as $EL6^m$. Although the metastable transformation probability of EL6 may be small, it takes a relatively short time to form the *E* state. Accordingly, a resultant time-dependent PPA signal shows the relatively fast quenching and enhancement to the *E* state as shown in Fig. 5 because of the overlapping of two photoinduced processes. The signal intensities of the *Q* state after the SLI of 0.9 eV on the *Q* and *E* states increase due to the fact that a part of EL6 already transforms to $EL6^m$. On the contrary, since all of EL6 transform to $EL6^m$ by the quenching-light illumination for a long time, the saturated signal intensities of the *E* state in three curves are the same as shown in Fig. 5.

In the present model, however, there still remain difficulties. As shown in Fig. 9, the broad band around 0.8–1.0 eV that appeared after the SLI of 0.9 eV on the *N* state is small in width and intensity compared with those of the *A* and *B* bands. Even after further illumination for longer than 30 min, there is no further increase in width and intensity of this broad band. According to the above discussions, the intensity and width of this broad band should be the same as those of the *A* and *B* bands. In addition, the PPA spectrum and time dependence are not influenced by the secondary light of 1.45 eV as shown in Figs. 7 and 8. Since we concluded that the SLI of 0.9 eV on the *E* state caused an optical recovery from $EL2^*$ to $EL2^0$, the PPA spectrum should increase in the higher photon-energy region above 1.1 eV. These imply that there is some correlation between the metastable transformation mechanisms of $EL6^m$ and $EL2^*$. A detailed explanation is not yet conclusive.

The present model is based on our previous report that the photoquenching and following enhancement of the PPA signal are due to the metastable transformation of EL2 and EL6, respectively.[7] We cannot present concrete evidence that the effects observed in the present paper are related to the presence and its metastable transformation of EL6. However, as is discussed in our previous paper, there are some indications for this fact. Although the chemical nature of EL6 is as yet unknown, this level is identified as a deep donor at 0.35 eV below CB. Its concentration is on the order of 10^{15} cm^{-3} in LEC-grown SI GaAs[6] and its electron photoionization spectrum has been reported to be similar to that of EL2 in the 0.7–1.5 eV spectral range.[10] These features indicate that the EL6 level plays an important role on the PPA signal generation mechanisms. Since the EL6 level has a much larger Frank-Condon shift of 0.6 eV than that of EL2 of 0.12 eV,[10] the nonradiative recombination probability of electrons involving EL6 may be larger than that of EL2. This fact explains our experimental result that the PPA signal intensities in the *E* state ($EL6^m$) is larger than that in the *N* (state EL2) despite the lower concentration of EL6 than EL2. The deep-level transient spectroscopy measurements should be carried out for the same samples to evaluate the EL6 concentration and proceed to further discussions.

IV. CONCLUSIONS

The effect of the secondary-light illuminations of $h\nu = 0.9$ and 1.45 eV on two photoinduced states, the quenched and the enhanced states, were investigated by using the PPA technique. We found that the secondary-light illumination of 0.9 and 1.45 eV on the quenched and enhanced state for 30 min at 80 K causes an optical recovery from $EL2^*$ to $EL2^0$. This is in agreement with the results reported by Manasreh and Fischer by using infrared optical-absorption measurements. We observed an appearance of the broad band at 0.8–1.0 eV after the secondary-light illumination for 30 min, which could not be observed in the PPA spectrum of the *N* state. The most important finding was that the PPA spectra after the secondary-light illumination of 0.9 eV on the *Q* and *E* states were the same in the whole photon-energy region. We concluded that the secondary-light illumination of 0.9 eV induced both an optical generation and dissociation of metastable state of EL6. The observed complex experimental results were explained by considering that the rates of transformation for these two processes were different under the secondary-light illumination of 0.9 eV.

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