

Photoacoustic signals of *n*-type GaAs layers grown by molecular-beam epitaxy on semi-insulating substrates

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(Received 24 February 1992; revised manuscript received 1 June 1992)

Piezoelectric photoacoustic (PA) measurements of molecular-beam-epitaxial (MBE)-grown GaAs layers were carried out in the temperature range from 90 to 290 K. A broad *D* band with a maximum near 1.3 eV and a sharp *Q* peak at 1.485 eV have been observed in the 90-K spectra. They vanish in a presence of secondary light illumination. By comparing with optical-absorption spectra, it is considered that the *D* band is due to electron transitions involving EL2 deep defect levels in the GaAs substrate. The PA signal is considered to be enhanced by the presence of the electric field at the interface between the MBE layer and the substrate. The *Q* peak is attributed to electron transitions from shallow acceptors such as carbon in GaAs. Observed photoinduced changes in the spectra are explained by a reduction of the electric field in the depletion region which is induced by optical carrier generation.

I. INTRODUCTION

Photoacoustic (PA) spectroscopy has recently been used to investigate physical properties of semiconductors for two reasons.^{1,2} One is that the PA measurement is a direct monitor of a nonradiative relaxation channel and therefore this may complement absorption and photoluminescence (PL) spectroscopic techniques. Wasa, Tsubouti, and Mikoshiba have succeeded in observing nonradiative deexcitation signals in PA spectra of CdS single crystals.³ The second reason is that the PA measurement is sensitive to a very small optical-absorption coefficient β in a highly transparent medium. Typical examples are investigations of midgap states in amorphous semiconductors. Caesar, Abkowitz, and Lin⁴ have detected values of β as low as 10^{-1} cm^{-1} for *a*-Se thin films with thicknesses less than $20 \mu\text{m}$ by using this technique. The presence of these two great advantages indicates that PA measurements should be a very useful tool for investigating deep levels in GaAs such as EL2.

Many investigations have been made in recent years toward understanding and identifying a native deep defect EL2 in bulk GaAs. Since EL2 serves to compensate the residual shallow acceptor levels by virtue of their midgap donor level, they are very important technologically for producing semi-insulating (SI) materials. From a more basic point of view, the EL2 has an ability to undergo a photoinduced transition from a normal to a metastable state at low temperature. The reverse recovery process from the metastable to the normal state occurs by thermal or optical stimulation. Although many investigations have been carried out to identify EL2 (Refs. 5 and 6) and some configurational-coordinate (CC) diagrams to interpret the electron transitions involving EL2 levels

have been proposed,⁷ the details are still controversial. Especially concerning the contribution of the nonradiative transitions, only a few experimental reports are available for GaAs crystals.

For the molecular-beam-epitaxial (MBE) grown GaAs buffer layers on bulk GaAs substrate, it has recently been shown that they can substantially reduce backgating, sidegating, and light sensitivity in metal-semiconductor field-effect-transistor devices.⁸ However, there are no extensive investigations of how dislocations or surface imperfections in the substrate influence the physical properties of the MBE-grown layers. Therefore, it becomes necessary to study the role of such dislocation- or defect-related levels including the EL2 in MBE-grown GaAs layers.

In the present paper, we report on the PA spectra of MBE-grown GaAs samples. We show that the observed broad band in the PA spectrum below band gap is due to a transition involving EL2 defect levels not in the epilayer but in the substrate. The electric field in the epilayer-substrate interface region may play a major role in the PA signal-generation mechanisms. We also show that PA spectroscopy is a useful tool for investigating deep states such as impurities or defects in semiconductors.

II. EXPERIMENTAL PROCEDURES AND RESULTS

The molecular-beam-epitaxial layers of GaAs were grown at a rate of $1.1 \mu\text{m/h}$ and at a temperature of 600°C . The *n*-type epilayer of $2\text{-}\mu\text{m}$ thickness was prepared by silicon doping and the electron concentration was estimated to be $3.8 \times 10^{16} \text{ cm}^{-3}$. The substrates

were semi-insulating (SI) $\langle 100 \rangle$ oriented GaAs wafers that had been grown by the liquid-encapsulated Czochralski (LEC) method. The samples for the PA measurements were cut to surface dimensions of about $0.5 \times 0.5 \text{ cm}^2$. The piezoelectric-transducer (PZT) detector was attached to the substrate side of the sample using a silver conducting paste. The incident light on the sample from a grating monochromator to measure the PA signal was mechanically chopped and always focused on the epilayer side. The experimental configuration of the sample and the detector is shown in the inset of Fig. 1. The sample was mounted on the cold finger of a liquid-nitrogen cryostat and then cooled down to 90 K in the dark. The probing light to obtain the PA spectrum was always kept sufficiently weak on the order of 10^{-5} W/cm^2 , so as not to generate any photoinduced changes of the spectra.⁹ Continuous secondary light with or without an IR transparent filter from a 50-W tungsten halogen lamp was used to investigate photoinduced effects. The secondary light was normally incident on the epilayer side of the sample. Chemical etching to remove the epilayer and to obtain a mirrorlike surface was carried out by immersing the sample in a $\text{H}_2\text{SO}_4(3):\text{H}_2\text{O}_2(1):\text{H}_2\text{O}(1)$ solution at 20°C . The etching rate was $40 \mu\text{m}/\text{min}$. The thicknesses of the nonetched and etched samples were 572 and $530 \mu\text{m}$, respectively. Details were reported previously.¹⁰

The photoacoustic spectra of the MBE-grown sample at room temperature (290 K) and at 90 K are shown in Fig. 1. The modulation frequency f was set at 77 Hz. Three characteristic features are obtained in the PA spectrum at 90 K. They are a broad band ranging from 0.8 to 1.44 eV, hereafter referred to as the *D* band, a peak at 1.466 eV (the *A* peak), and a stronger peak at 1.485 eV (the *Q* peak). The *Q* peak is about three times larger in height than the *A* peak. Since the energy gap of GaAs is 1.508 eV at 90 K,¹¹ the band and the peaks are considered to originate from localized states in the band gap. When the sample was heated up to 290 K, the spectrum shifted to the lower-energy side. The *D* band and the *Q* peak become smaller compared with the *A* peak. At room temperature, the *Q* peak has an intensity eight

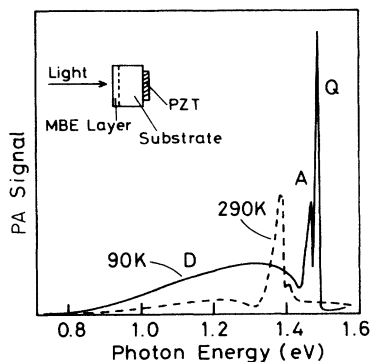


FIG. 1. The photoacoustic (PA) spectrum of the MBE-GaAs layer at 90 K (solid curve). The modulation frequency was set at 77 Hz. The broad band (*D*) and the peaks at 1.466 (*A*) and 1.485 eV (*Q*) are observed. The spectrum at 290 K is denoted by a dashed curve.

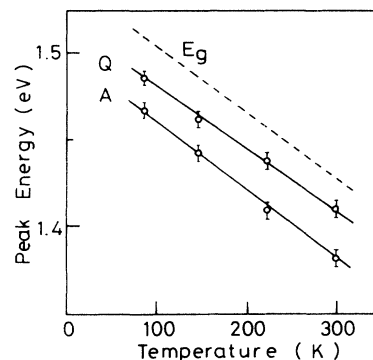


FIG. 2. The temperature variation of the *A* and *Q* peaks. The energy gap E_g is also shown by a dashed line. The straight lines were obtained by least-squares fits.

times smaller than that of the *A* peak. In Fig. 2, the temperature variations of the peak energies for the *A* and *Q* peaks are shown by the solid lines. The variation of the energy gap is shown by the dashed line.¹¹ The peaks shift linearly to lower energy with an increase in temperature. The slope for the *Q* peak ($3.7 \times 10^{-4} \text{ eV/K}$) is slightly smaller than that of the others ($4.0 \times 10^{-4} \text{ eV/K}$).

Figure 3 shows the effect of secondary light illumination on the PA spectra at 90 K. The dashed curve is for the spectrum in the dark (the same as that in Fig. 1). The solid curve is for that under white light illumination. The *D* band and the *Q* peak vanished completely, but the intensity of the *A* peak was almost unchanged by the illumination. When the secondary light was switched on, the PA signal intensity near 1.3 eV suddenly decreased. When the light was switched off, it took a few sec for the PA signal to recover to the values in the dark. When an IR transparent filter was used to cut the secondary light of wavelength smaller than 950 nm, a similar spectrum was observed. It was shown by the dotted-dashed curve in the figure. The *Q* peak completely vanishes, but the reduction of the *D* band is not drastic in this case. A broad but small band remains.

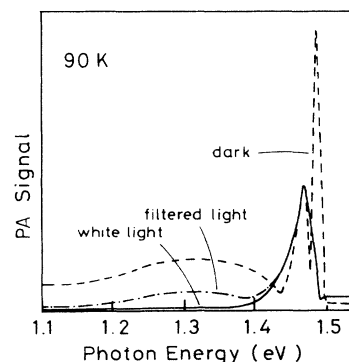


FIG. 3. The effect of the secondary light on the PA spectra at 90 K (solid curve). The light was filtered (dotted-dashed curve) by an IR transparent filter or unfiltered (solid curve). The *D* band and the *Q* peak are reduced significantly by the illumination. The spectrum in the dark is given by the dashed curve for comparison.

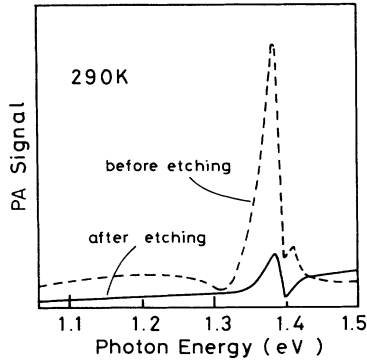


FIG. 4. The PA spectrum of a chemically etched sample, where the MBE layer was completely removed. The Q peak vanishes and the D band becomes weak. The edge of the plateau at 1.43 eV was assigned to the energy gap E_g . The spectrum of a nonetched sample is also shown (dashed curve).

Since the MBE-GaAs layers were grown on SI-GaAs substrates, the contributions from both epilayer and substrate should be simultaneously considered for PA signal generation. To separate these contributions, the PA spectrum measurements have been also carried out for a chemically etched sample. The epilayer of $2\text{-}\mu\text{m}$ thickness was completely removed. The observed spectrum at room temperature is shown in Fig. 4 by the solid curve. The PA spectrum for the sample before etching is shown by the dashed curve to facilitate the comparison. Since the change of sample thickness by etching was less than 10%, there should be no effect on the PA spectra by the difference in sample thicknesses. The overall features of the etched sample are very similar to those of the SI-GaAs wafers reported earlier.¹⁰ The Q peak does not occur and the D band becomes more gradual. The edge of the plateau at 1.43 eV in the spectrum of the etched sample has been assigned to the band gap of GaAs at room temperature.

III. DISCUSSION

In the photoacoustic spectra of MBE-grown samples, the A and Q peaks and the broad D band are observed in the whole temperature region from 290 to 90 K (Fig. 1). PA measurements on LEC-grown SI and high-resistivity n -type GaAs wafers were carried out previously.^{10,12} A continuous broad band near 1.35 eV and a peak at 1.383 eV were also observed at room temperature. They correspond to the D band and the A peak, respectively, in the present study. We had concluded there that the D band was due to electron transitions involving EL2 deep defect levels. For the 1.383-eV peak, the origin was considered to be dislocation related. A possibility that the A peak is an apparent one expected from the proposed model¹³ was discounted by comparing the experimental results with calculated PA spectra. In our earlier investigations for the PA spectra of SI-GaAs wafers,^{10,12} no peak corresponding to the Q peak appeared. Therefore, the appearance of the Q peak and the enhancement of the D band, which shows a distinctive maximum, are considered to be characteristic features for the present MBE-grown sam-

ples. This proposition will be confirmed by the PA spectrum for the etched sample in the next paragraph. In the present paper, we have focused our discussions on the D band and the Q peak.

The MBE-grown GaAs layer was deposited on the SI GaAs substrate wafer in the present study, and the PA signal was detected by a PZT transducer on the rear side of the substrate, as shown in the inset of Fig. 1. Thus the PA signals generated in the epilayer and substrate regions could be simultaneously detected. To distinguish the PA signal of the epilayer from the substrate contribution, the PA measurements were also carried out for a chemically etched sample, where the epilayer was completely removed. In the PA spectra of the etched sample (Fig. 4), the D band decreases drastically and the Q peak vanishes. The observed spectrum for the etched sample is the same as that reported earlier for SI GaAs wafers.^{10,12} Therefore, we consider that enhancement of the D band and appearances of the Q peak are characteristic features of the epilayer itself or of the interface between the epilayer and the substrate. We discuss PA signal-generation mechanisms for the D band and the Q peak below.

A. The D band

In the PA spectrum at 90 K in Fig. 1, the D band has a broad maximum near 1.3 eV. Comparing with the PA spectra of SI-GaAs (Ref. 10) or the etched sample in Fig. 4, it is reasonable to conclude that the D band becomes predominant for the MBE samples. For the substrate GaAs wafers, the PA signal monotonically increases with incident photon energy and merges into the tail of the A peak. A similar continuous absorption band below E_g has also been observed for SI LEC-grown GaAs by IR-absorption measurements at 9 K by Fischer,⁷ who concluded that this absorption is due to EL2 deep levels. Since the absorption coefficient β due to the EL2 defects is expected to be lower than 10 cm^{-1} , the product βL becomes 0.5 when the sample thickness is about $500\text{ }\mu\text{m}$. This is in the region where a linear dependence of the PA signal on β would be expected.¹³ This means that the observed signal in the PA spectrum below the A peak directly reflects the absorption spectrum. Therefore, we first consider that the broad band below 1.35 eV in the PA amplitude spectrum (the D band) is due to transitions involving EL2 centers from the similarity with the optical-absorption spectrum. The EL2 signals in the MBE samples are enhanced by the presence of the epilayer. Detailed PA signal-generation mechanisms for the enhancement are given in the following.

Since the intensity of the D band is stronger in the MBE sample than in the bulk SI sample, we first assume that the PA signal is generated by EL2 in the epilayer, which has a thickness of $2\text{ }\mu\text{m}$. A continuous absorption band has been observed for a MBE-grown GaAs layer by infrared-absorption measurements at 9 K by Manasreh *et al.*⁸ They have grown epilayers at a very low substrate temperature of about 200°C . Unlike the epilayers grown at around 600°C , they have pointed out that the low-temperature-grown layer contains deep defects with concentrations as high as 10^{19} cm^{-3} . They also concluded

that these defects are very similar to EL2 by considering characteristic features of the photoquenching and subsequent thermal or optical recovery from the metastable states. Since our epilayers were grown at the relatively high temperature of 600 °C, the concentration of EL2 levels should be very low, and it would not be expected that they could be detected. A typical EL2 concentration in normal MBE buffer layers has been reported to be on the order of 10^{12} cm^{-3} , as determined by a deep-level transient spectroscopy (DLTS) analysis.¹⁴ This is much smaller than the EL2 concentration of 10^{16} cm^{-3} in SI-LEC samples.⁵ Furthermore, the thickness of the substrate is more than 100 times larger than that of the epilayer. These features indicate that the contribution from the substrate to the PA signal should be dominant. Accordingly, it is not likely that the *D* band is due to the EL2 levels in the epilayer.

As discussed above, the *D* band is strongly enhanced in the MBE sample and reduces when the epilayer is completely removed. Since this band is not due to EL2 in the epilayer discussed above, we alternatively propose a new model to explain the experimental results. The *D* band may be generated by nonradiative recombination of electrons through interface states between the epilayer and the substrate. These electrons are assumed to be optically excited from EL2 levels in the bulk substrate, where the EL2 concentration is considerably higher than that in the epilayer. The electric field in the depletion region near the interface may enhance the nonradiative transitions. We next discuss this model in more detail.

Since the epilayer is silicon doped and the electron concentration n is $3.8 \times 10^{16} \text{ cm}^{-3}$, the Fermi level is located very near the conduction band. Using a value of $4.7 \times 10^{17} \text{ cm}^{-3}$ for the effective density of states in the conduction band at room temperature,¹¹ the Fermi level is estimated to be about 0.065 eV below the bottom of the conduction band. The EL2 donor activation energy was estimated to be 0.76 eV below the conduction band by the Hall measurements⁷ and, typically, the Fermi level in a SI substrate is located slightly above the EL2 level. Therefore, when the epilayer-substrate junction is fabricated, a depletion region with an accompanying electric field is formed at the interface between the epilayer and the substrate. The depletion-region width is determined from Poisson's equation by using the electron carrier concentration n in the *n*-type epilayer and the below-midgap acceptor density N_T^- in the SI substrate. Since N_T^- is usually less than 10^{16} cm^{-3} in SI GaAs, the depletion region is mainly extended into the substrate region. A schematic band diagram is shown in Fig. 5. The EL2 level in the epilayer is not shown because of its expected low concentration. At the interface between the epilayer and vacuum, surface states which take up carriers and leave a depletion region are assumed. Intrinsic light absorption in the sample produces electron-hole pairs. Pairs produced in the depletion region or within a diffusion length in the substrate side will eventually be separated by the electric field. Electrons will drift to the epilayer side; otherwise, holes will drift to the substrate side.

It was shown that interface states between the epilayer and the substrate influence the formation of the depletion

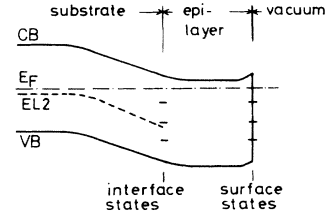


FIG. 5. Schematic band diagrams near the epilayer-substrate interface. EL2 levels are shown in the substrate region. Electrons drift to the interface and recombine nonradiatively with the interface states.

region. Since the depletion region induced by these interface states is believed to degrade severely the performance of electronic and optical devices, investigations of the MBE growth technique have been extensively carried out. Interface states produced during MBE growth are mainly generated by dislocations, surface roughness, or surface contamination, such as oxygen and carbon atoms of the substrate materials. In the conventional MBE technique, it is not easy to remove these interface states, especially when the LEC GaAs wafer was used as the substrate material. Look, Stuzze, and Evans¹⁵ have reported that interface states as high as $1.2 \times 10^{12} \text{ cm}^{-2}$ were present when a MBE *n*-type GaAs layer of $n = 1.7 \times 10^{17} \text{ cm}^{-3}$ was grown on a SI GaAs substrate. Therefore, it is reasonable to expect the presence of such interface states which can trap optically excited electrons or holes.

The probing light for the PA signal generation sufficiently penetrates deep into the bulk substrate. In our model, the optically generated electrons from the EL2 levels in the substrate drift to the epilayer-substrate interface. They eventually recombine with the interface states nonradiatively and cause the stronger *D* band in the PA spectrum. An additional nonradiative transition path becomes available by the presence of the epilayers. The electric field in the depletion region and the presence of the interface states are responsible for the PA signal-generation mechanism.

We next consider the effect of secondary light illumination on the *D* band. It has been reported that the EL2 absorption band is photoquenched by secondary light illumination, most effectively at a photon energy of 1.12 eV. The recovery of the quenched band can be accomplished by thermal or optical treatment.⁶ These experimental features have been well interpreted by considering that photoquenching occurs when the EL2 defect is transformed from its normal to a metastable state. Our observed photoinduced effect seems to be explained by photoquenching of the EL2 levels. When the secondary light is applied, the electrons in the normal states are excited to the metastable states. Thus, the effective number of carriers that are excited by the probing light and then cause the PA signal decreases. When the light is off, the carriers in the metastable states return to the normal states by thermal processes. Thus, the PA signal again increases up to its value in the dark. The thermal recovery process of the metastable EL2 defects in GaAs

has been analyzed by Parker and Bray.⁶ They have measured the near-band-edge absorption spectrum of SI GaAs wafers. The absorption signal below E_g is completely photoquenched below 110 K and is completely recovered above about 120 K.

The photoinduced effect in Fig. 3 was observed at 90 K in our experiment. If the carriers were optically excited to the metastable states by the secondary light illumination, they should recover to the normal states when the light was switched off. Otherwise, we cannot explain our experimental results. Since the measuring temperature of 90 K is not sufficient to cause the thermal recovery of the electrons from the metastable states, alternative explanations for the recovery process should be considered.

Another possible explanation for the photoinduced effect of our sample is that the strength of the electric field in the depletion region decreases due to the generation of free carriers under secondary light illumination. The optically generated electrons and holes in the depletion region are swept to the epilayer side and to the bulk substrate side, respectively. Since we considered that the PA signal is enhanced by the presence of the electric field, the *D* band should decrease with the decrease of the electric field. This is the case of Fig. 3. When the secondary light is filtered, where only an extrinsic light below the band gap is available, electron excitation to the conduction band may be dominant. However, when white light is illuminated, electrons and holes are simultaneously generated. The latter drastically reduces the electric field in the depletion region. Thus, illumination by the white light more effectively reduces the *D* band in the spectrum, as shown in Fig. 3.

B. The *Q* peak

As discussed before, the observed *Q* peak at 1.485 eV (90 K) is also a specific feature of the MBE sample. The peak intensity decreases significantly with the increase in temperature. And when the secondary light is applied, the peak disappears, as in the case for the *D* band. We consider the possibility that the *Q* peak is due to nonradiative recombination of free carriers excited from shallow impurity states. Electrons in the conduction band drift to the epilayer-substrate interface by the presence of the electric field in the depletion region and nonradiative recombination takes place at the interface states. If the excited carriers are holes, they drift to the substrate side and do not contribute to the PA signal. Thus we believe that the electrons which are excited from shallow acceptors cause the *Q* peak in the PA spectrum. The temperature variation of the *Q* peak energy seems to support this proposition.

The *Q* peak intensity also decreases under secondary light illumination, as shown in Fig. 3. This is also explained by the reduction of the electric field in the depletion region, as discussed in Sec. III A. The presence of shallow acceptors causes the *Q* peak, and the energy difference of the *Q* peak and energy gap (E_g) at 90 K is estimated to be as large as 25 meV. These results suggest that the *Q* peak is due to shallow acceptors, such as carbon atoms in arsenic lattice sites. Their activation energy

is reported at 20–25 meV (Ref. 16) and agrees well with our data. The presence of residual carbon atoms in GaAs is probable.

Similar photoinduced changes in the transverse acoustic-electric voltage (TAV) spectra were observed by Davari and Das for Cr-doped SI (GaAs).¹⁷ This technique relies on the interaction of the electric field accompanying a surface acoustic wave on a piezoelectric material with free carriers at the semiconductor surface. The observed voltage is proportional to $(n\mu_e - p\mu_h)$, where n and p are the carrier concentrations and μ_e and μ_h are the mobilities for electrons and holes, respectively. In our proposed model, electrons and holes drift to the opposite sides in the depletion region due to the presence of the electric field. This suggests that the principles for the TAV signal-generation mechanism may be similar to those of the PA spectra in the present case. The possibility that our *D* band and the *Q* peak have the same origins as those in the TAV spectrum still remains. More thorough experiments and analysis are necessary for further consideration of this point.

It is necessary to comment here on the invariance of the *A* peak height under secondary light illumination, as shown in Fig. 3. The *A* peak arises from the bulk SI GaAs substrate. Since we have considered that the PA signal is mainly due to optically excited electrons in the substrate region, the photoinduced effect for the *A* peak should be the same as those for the *D* band and the *Q* peak. However, supposing that the *A* peak is due to donor states in the substrate, the invariance of the *A* peak height may be explained. In this case, the optically generated carriers are holes. Holes drift to the substrate bulk side by the electric field in the depletion region. Therefore, they are not influenced by the presence of the epilayer. They recombine nonradiatively within the substrate material and cause the PA signal. Thus the presence of the epilayer may not drastically affect the features of the *A* peak in the spectra.

IV. CONCLUSIONS

We have observed two characteristic features in the PA spectra of MBE-grown GaAs samples. They are a broad *D* band around 1.3 eV and a sharp *Q* peak at 1.485 eV at 90 K. We have concluded that the PA signals are caused by nonradiative recombination of the electrons from the extrinsic levels in the substrate region. They are from the EL2 deep defect level for the *D* band and from shallow acceptor levels, such as carbon on arsenic lattice sites for the *Q* peak. The excited electrons drift to the epilayer-substrate interface due to the electric field in the depletion region and then recombine through the interface states. The interface states may be due to extrinsic surface states of the substrate material which could not be removed completely during the MBE growth procedures.

A distinct photoinduced reduction of the *D* band by secondary light illumination could be observed in the present study. If we suppose that this effect is due to the photoquenching of the electrons in the EL2 levels to the metastable states, highly efficient recovery processes should be simultaneously taken into account. This is not

the case in the present studies. Therefore, we suppose that the observed photoinduced reduction of the *D* band is explained by a decrease of the electric field in the depletion region due to the generation of free carriers. We feel that our considerations for the origin of the *D* band and the *Q* peak are reasonable as a first approximation.

ACKNOWLEDGMENTS

We wish to thank Dr. O. Otsuki and Dr. M. Ozeki of Fujitsu Laboratory Co. Ltd. for preparing the MBE-grown GaAs samples.

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