Defect density measurements of low temperature grown molecular beam epitaxial GaAs by photothermal deflection spectroscopy

M. H. Chan and S. K. So[a)]
Department of Physics, Hong Kong Baptist University, 224 Waterloo Road, Kowloon, Hong Kong

K. T. Chan
Department of Electronic Engineering, The Chinese University of Hong Kong, Shatin, Hong Kong

F. G. Kellert
Microwave Technology Division, Hewlett-Packard Company, 1412 Fountaingrove Parkway, Santa Rosa, California 95403

(Received 15 November 1994; accepted for publication 29 May 1995)

The subgap optical absorption of GaAs layers grown by low temperature molecular beam epitaxy is measured by photothermal deflection spectroscopy (PDS). The absorption increases as the growth temperature decreases at a fixed wavelength. Defect densities evaluated from the absorption spectra and the known absorption cross sections are between 10^{18} and 10^{19} cm^{-3}. It is shown that complementary PDS phase spectra can be used to separate the absorption of the epitaxial layers from the bulk. © *1995 American Institute of Physics.*

Epitaxial layers of GaAs grown by low temperature molecular beam epitaxy (LT-MBE) have recently been shown to possess very useful properties for device fabrication. For example, these materials have been used as a buffer layer to suppress backgating and sidegating in field effect transistors.[1] Photoconductive devices grown from LT-MBE GaAs exhibit very fast response time.[1] It is now generally believed that these special properties of LT-MBE GaAs are due to an excess amount of As present during epitaxial growth. High concentration of point defects is found in LT-MBE GaAs and is thought to be associated with the excess As in forms of As antisites (As_{Ga}), As interstitials, As precipitates after annealing, or their complexes. An understanding of the nature of these defects will, therefore, provide useful insights for improving the performance of devices constructed from these materials.

Several techniques have been used to study the defects in LT-GaAs grown by MBE including electron paramagnetic resonance,[2–4] optical absorption,[5,6] Hall effects,[7,8] photoluminescence,[9,10] and deep-level transient spectroscopy.[11,12] From optical absorption measurements,[5,6] it was discovered that the shapes of the absorption spectra of LT-GaAs closely resemble those of intrinsic GaAs grown by the liquid-encapsulated Czochralski (LEC) method. Since As antisite defects are present in both LT-MBE and LEC materials, defects in both cases should have very similar properties. The majority of defects in LEC grown GaAs are now generally accepted to be due to EL2 defects ($\sim As_{Ga}$), and therefore, it was proposed that defects in LT-GaAs are also EL2-like.[5,6] Some of the key parameters for evaluating the EL2-like defect concentrations are the photoionization cross sections for neutral defects (σ_n) and positive defects (σ_p). For GaAs, the photoionization cross sections for both neutral and positive As antisites have been evaluated using photocapacitance measurements.[13,14] In this study, we demonstrate for the first time that it is possible to use photothermal deflection spectroscopy (PDS), a very sensitive technique for measuring optical absorption, to evaluate defect densities in LT-GaAs using known photoionization cross sections for EL2 in GaAs.

The GaAs MBE layers (1 μm thick) were grown on a 100 Å thick buffer layer of AlAs using LEC GaAs as the substrate. The MBE layers were subsequently annealed at 620 °C for 25 min in the MBE chamber under an As-rich condition. The AlAs buffer layer has a relatively large bandgap energy (2.2 eV) and can be considered to be optically transparent in this study. GaAs epilayers grown at different substrate temperatures (200, 225, 250, 300, and 585 °C) were investigated by PDS. The principle of operation of PDS is well documented.[15] The measurements were carried out with a standard setup consisting of a 1 kW Xe arc lamp and a 1/4 m grating monochromator (Oriel) with selective long-pass filters as a tunable light source. The pump beam was modulated by a chopper before irradiating on the sample. Carbon tetrachloride was used as a deflecting fluid. A Uniphase HeNe laser was directed parallel to the layer surface as the probe laser. A quadrant cell (United Detector Technology) was used as the position sensor for monitoring the photothermal deflection signal of the probe beam. The output of the detector was fed into a lock-in amplifier (Standard Research Model SR830) for phase sensitive measurements. All PDS spectra were normalized to the incident power of the pump beam.

Results of normalized PDS spectra on MBE grown GaAs are shown in Fig. 1. The spectra were calibrated by transmission measurements using a multilayer model.[6] It is observed that the subgap absorption, which correlates closely to the defect density, increases as the substrate temperature decreases at a fixed wavelength. The EL2-like defect densities in the MBE layer, $[As_{Ga}]^0$ and $[As_{Ga}]^+$, can be calculated accurately with the known electron and hole photoionization cross sections σ_n and holes σ_p, respectively, according to the equation

[a)]Author to whom correspondence should be addressed. Electronic mail: skso@ctsc.hkbc.hk

 0003-6951/95/67(6)/834/3/$6.00 © 1995 American Institute of Physics

Copyright ©2001. All Rights Reserved.

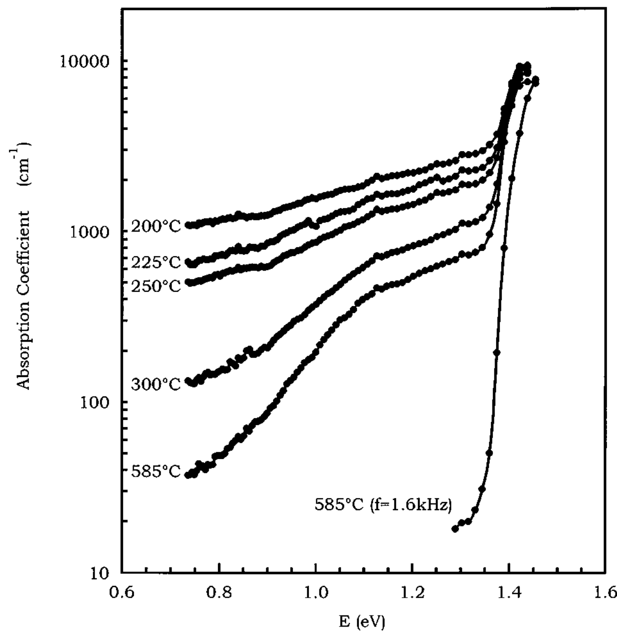

FIG. 1. The absorption spectra measured by PDS for MBE layers grown at different substrate temperatures. The chopping frequencies are 13 Hz unless otherwise indicated.

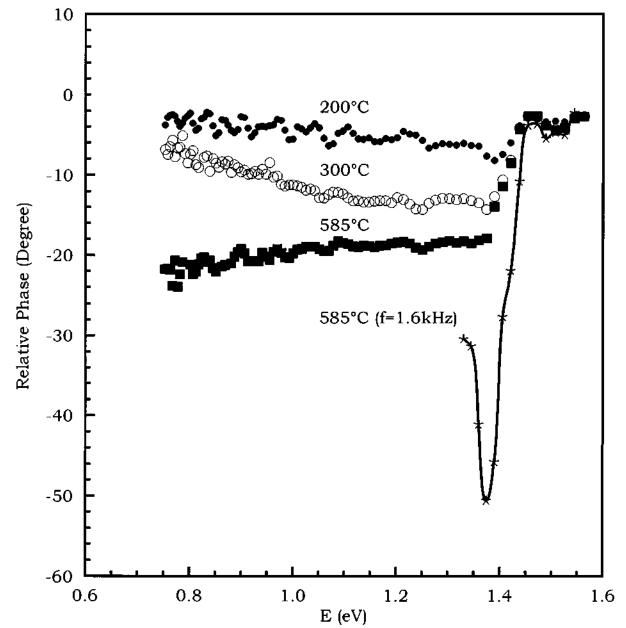

FIG. 2. The PDS phase spectra for MBE layers grown at different substrate temperatures (some of the phase spectra are not shown for clarity). The phase spectra were taken simultaneously with the PDS spectra shown in Fig. 1. The chopping frequencies are 13 Hz unless otherwise indicated.

$$\alpha(\lambda)=\sigma_n(\lambda)[\mathrm{As_{Ga}}]^0+\sigma_p(\lambda)[\mathrm{As_{Ga}}]^+, \quad (1)$$

where $\alpha(\lambda)$ is the absorption coefficient at wavelength λ for the MBE layer. Since the shapes of the absorption spectra in both the MBE layer and the substrate are very similar and $\mathrm{As_{Ga}}$ is involved in both cases,[6] it can be assumed that the photoionization cross sections for the substrate can be used in (1) for evaluating the defect concentrations. In addition, PDS is known to be sensitive to the surface rather than the bulk.[15,16] Hence, for LT-MBE GaAs, the absorption due to the GaAs epilayer is expected to be larger than that of the substrate due to high defect density of the epilayer (about 10^{18} cm^{-3} for annealed LT-MBE GaAs grown at 200 °C[1,5,6]). Therefore, the PDS signal should be dominated by the epilayer. Based on the assumptions above, the defect densities for the epilayers, $[\mathrm{As_{Ga}}]^0$ and $[\mathrm{As_{Ga}}]^+$, can be calculated from Eq. (1) using the measured absorptions at two different wavelengths. For LEC GaAs, the cross sections are $\sigma_n(1.1\ \mu\mathrm{m})=9.07\times10^{-17}$, $\sigma_n(1.2\ \mu\mathrm{m})=4.8\times10^{-17}$, $\sigma_p(1.1\ \mu\mathrm{m})=3.2\times10^{-17}$, and $\sigma_p(1.2\ \mu\mathrm{m})=4.72\times10^{-17}$ cm^2.[13,14] Results of the defect concentrations using these known values of absorption cross sections are shown in Table I for epilayers grown between 200 and 300 °C. The errors are estimated from the assumption of $\pm5\%$ error in the cross sections and are dependent on the photothermal deflection signal. Total defect concentrations are in the range of $10^{18}-10^{19}$ cm^{-3}. The total defect density calculated according to the above method appears to be abnormally high (5.5×10^{18} cm^{-3}) for the MBE layer grown at 585 °C, in sharp contrast to reports that the MBE layer grown at 580–600 °C can produce $\sim10^{13}$ cm^{-3} shallow donor and acceptor concentrations, with the deep trap concentrations smaller by at least a factor of 10.[17]

The apparent contradiction above can be explained by a careful consideration of the PDS phase spectra in Fig. 2. For the MBE layers grown at 200, 225, 250, and 300 °C, as both the MBE layer and the bulk become more transparent (just below the gap energy), the effective center of absorption moves away from the surface and causes a sudden phase lag in the PDS signal. Below the band-gap energy, the phase lag becomes smaller for smaller photon energies because the center of absorption moves to the front surface where the absorption is dominated by the MBE layer. Similar phase motion has been observed previously by Zammit *et al.*[18,19]

The PDS phase spectrum for the 585 °C sample (ob-

TABLE I. Summary of the EL2-like defect densities of annealed MBE layers grown at different substrate temperatures obtained from Eq. (1) and the optical sum rule. The chopping frequency employed is 13 Hz.

Substrate temperature (°C)	$[\mathrm{As_{Ga}}]^0$ ($\times10^{19}$ cm^{-3})	$[\mathrm{As_{Ga}}]^+$ ($\times10^{19}$ cm^{-3})	Total defect density $[\mathrm{As_{Ga}}]^0+[\mathrm{As_{Ga}}]^+$ ($\times10^{19}$ cm^{-3})	Total defect density (from sum rule) $\times10^{18}$ cm^{-3})	$\frac{[\mathrm{As_{Ga}}]^+}{[\mathrm{As_{Ga}}]^0+[\mathrm{As_{Ga}}]^+}$
200	1.7 ±0.1	1.8 ±0.1	3.5 ± 0.2	1.9	0.51
225	1.4 ±0.1	1.2 ±0.1	2.6±0.2	1.5	0.46
250	1.2 ±0.1	0.81±0.06	2.0 ± 0.2	1.2	0.40
300	0.72±0.05	0.21±0.02	0.93± 0.06	0.68	0.23

Copyright ©2001. All Rights Reserved.

tained at a chopping frequency $f = 13$ Hz) also shows a similar phase shift just below the band gap. However, the phase lag becomes larger for smaller photon energies. This phase dependence shows that the effective center of absorption shifts gradually to the bulk and remains there. The phase spectrum ($f = 13$ Hz) suggests that the corresponding absorption spectrum for the 585 °C sample (f=13 Hz) in Fig. 1 is a measure of the average absorption of the entire sample rather than the MBE layer alone. Since the modulated heating from the pump beam is spread over a distance of the order given by the thermal diffusion length (μ_T), the contribution to the PDS signal due to the MBE layer can be enhanced by reducing μ_T which is given by $\mu_T = [D/(\pi \times f)]^{1/2}$, where D is the thermal diffusivity (0.003 $\text{cm}^2\,\text{s}^{-1}$ for GaAs)[20] and f is the chopping frequency. At a chopping frequency of 13 Hz, $\mu_T \approx 86$ μm. The major contribution to the absorption is due to the substrate because MBE layer grown at 585 °C is known to have relatively few defects and therefore should be essentially transparent below the band gap. At higher chopping frequencies, the contribution of the epilayer to the PDS signal is expected to be enhanced because μ_T is reduced. To support this argument, PDS was performed again at a chopping frequency of 1.6 kHz where $\mu_T \approx 8$ μm. The measured absorption for the 585 °C sample is significantly reduced (Fig. 1). Simultaneously, the PDS phase spectrum exhibits the characteristic phase motion where the phase lag increases sharply just below the band gap followed by a smaller phase lag (Fig. 2). On the other hand, PDS measurements performed at a chopping frequency of 1.6 kHz on other MBE layer are similar to those obtained at a chopping frequency of 13 Hz. These measurements confirm that PDS only measures the absorption of the MBE layers grown at 200, 225, 250, and 300 °C at a chopping frequency of 13 Hz. It is more desirable but difficult to reduce the thermal diffusion length to 1 μm as a chopping frequency of 10^5 Hz is required.

The total defect concentration for the annealed MBE layer grown at 200 °C is about an order of magnitude higher than the value (3×10^{18} cm^{-3}) observed by Look *et al.* for both absorption and Hall effect measurements.[1,5–7] Also, the compensation ratio of deep acceptors $[\text{As}_{\text{Ga}}]^+$ to total defect density ($[\text{As}_{\text{Ga}}]^0 + [\text{As}_{\text{Ga}}]^+$) for the MBE layer grown at 200 °C is about 0.5 (Table I) whereas about 0.1 is observed by Look *et al.*[6,7] The discrepancy may be attributed to different annealing conditions. In Look's study, post-growth anneals on the LT-MBE GaAs were performed in a flowing inert gas. In our study, post-growth anneals on the MBE GaAs at 620 °C were performed *in situ* under arsenic overpressure.

The EL2-like defect can also be estimated from the optical sum rule which has been applied to estimate the concentration of dangling bonds in *a*-Si:H and the defect density of semi-insulating GaAs.[21,22] The defect density N is given by

$$N = \frac{cn\mu}{2\pi^2\hbar^2 e_s^{*2}} \int \alpha(E)\,dE, \tag{2}$$

where c is the speed of light, n ($\approx$3.5 for GaAs) is the refractive index, μ is the electron effective mass, $\alpha(E)$ is the absorption coefficient at photon energy E, and e_s^* is the effective charge of the defect. With the Szigeti-type local field correction, the effective charge of the defect for a homogeneous medium is given by[23]

$$e_s^{*2} = \tfrac{1}{9}(n^2+2)^2 e^2 f_{oj}, \tag{3}$$

where e is the electron charge, and the oscillator strength f_{oj} is assumed to be unity. The defect densities calculated from the sum rule are also shown in Table I. Results from the sum rule are significantly smaller than those obtained from Eq. (1). The large difference in the defect densities can be attributed to the uncertainty in estimating the effective charge of the defects. Brodsky *et al.*[23] have applied the local field correction to calculate e_s^*. An order of magnitude variation for e_s^* was obtained depending on whether the defect was embedded in a homogeneous or an inhomogeneous medium. The uncertainty in evaluating the effective charge is the major error in determining the defect densities from the sum rule.

Support of this research by the Research Grant Council under Grant No. RGC/93-94/12 (HKBC 107/93E) is gratefully acknowledged.

[1] David C. Look, Thin Solid Films **231**, 61 (1993).
[2] M. Kaminska, E. R. Weber, Z. Liliental-Weber, R. Leon, and Z. U. Rek, J. Vac. Sci. Technol. B **7**, 710 (1989).
[3] H.-J. Sun, G. D. Watkins, F. C. Rong, L. Fotiadis, and E. H. Poindexter, Appl. Phys. Lett. **60**, 718 (1992).
[4] K. Krambrock, M. Linde, J. M. Spaeth, D. C. Look, D. Bliss, and W. Walukiewicz, Semicond. Sci. Technol. **7**, 1037 (1992).
[5] M. O. Manasreh, D. C. Look, K. R. Evans, and C. E. Stutz, Phys. Rev. B **41**, 10272 (1990).
[6] D. C. Look, D. C. Walters, M. Mier, C. E. Stutz, and S. K. Brierley, Appl. Phys. Lett. **60**, 2900 (1992).
[7] D. C. Look, D. C. Walters, M. O. Manasreh, J. R. Sizelove, C. E. Stutz, and K. R. Evans, Phys. Rev. B **42**, 3578 (1990).
[8] D. C. Look, D. C. Walters, G. D. Robinson, J. R. Sizelove, M. G. Mier, and C. E. Stutz, J. Appl. Phys. **74**, 306 (1993).
[9] I. Ohbu, M. Takahama, and K. Hiruma, Appl. Phys. Lett. **61**, 1679 (1992).
[10] P. W. Yu, D. C. Reynolds, and C. E. Stutz, Appl. Phys. Lett. **61**, 1432 (1992).
[11] C. E. C. Wood, J. Woodcock, and J. J. Harris, Inst. Phys. Conf. Ser. **45**, 29 (1979).
[12] R. A. Stall, C. E. C. Wood, P. D. Kirchner, and L. F. Eastman, Electron. Lett. **16**, 171 (1980).
[13] P. Silverbery, P. Omling, and L. Samuelson, Appl. Phys. Lett. **52**, 1689 (1988).
[14] S. K. Brierley and D. S. Lehr, Appl. Phys. Lett. **55**, 2426 (1989).
[15] W. B. Jackson, N. M. Amer, A. C. Boccara, and D. Fournier, Appl. Opt. **20**, 1333 (1981).
[16] Andreas Mandelis, J. Appl. Phys. **54**, 3404 (1983).
[17] H. Sakaki, in *III-V Semiconductor Materials and Devices,* edited by R. J. Malik (North-Holland, Amsterdam, 1989), p. 217).
[18] U. Zammit, F. Gasparrini, M. Marinelli, R. Pizzoferrato, F. Scudieri, and S. Martellucci, J. Appl. Phys. **69**, 2577 (1991).
[19] U. Zammit, M. Marinelli, and R. Pizzoferrato, J. Appl. Phys. **69**, 3286 (1991).
[20] L. Luciani, U. Zammit, M. Marinelli, R. Pizzoferrato, F. Scudieri, and S. Martellucci, Appl. Phys. A **49**, 205 (1989).
[21] N. M. Amer and W. B. Jackson, in *Semiconductors and Semimetals* edited by J. I. Pankove (Academic, New York, 1984), Vol. 21B, 83–112.
[22] U. Zammit, F. Gasparrini, M. Marinelli, R. Pizzoferrato, F. Scudieri, S. Martellucci, Appl. Phys. A **52**, 112 (1991).
[23] M. H. Brodsky, Manuel Cardona, and J. J. Cuomo, Phys. Rev. B **16**, 3556 (1977).

Copyright ©2001. All Rights Reserved.