

Photoluminescence from heteroepitaxial (211)B CdTe grown on (211)B GaAs by molecular beam epitaxy

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Low temperature (~ 5 K) photoluminescence spectroscopy was performed on undoped CdTe epilayers grown by molecular beam epitaxy on (211)B oriented bulk GaAs substrates at substrate temperatures ranging from 230 to 275 °C. The emission spectra from all samples studied contained evidence of the diffusion of gallium and arsenic atoms from the substrate. A broad, low amplitude emission band observed at 1.594 eV was related to the Ga_{Cd} donor level in CdTe. Donor-acceptor pair recombination observed at 1.51 eV was due to the substitutional Ga_{Cd} donor and As_{Te} acceptor. The level of compensation in the CdTe layers was determined from the energy shift of the donor-acceptor emission peak with excitation power, with the lowest degree of compensation observed in a sample grown at 230 °C. In addition, a bright emission peak was observed at 1.47 eV. This peak, which had been observed previously in homoepitaxial and heteroepitaxial growth of CdTe, was related to electron-hole recombination of a structural defect in the CdTe/GaAs epilayers with an electronic binding energy of ~ 130 meV.

I. INTRODUCTION

CdTe is a direct-band-gap semiconductor that serves as a nearly lattice-matched substrate for growth of Hg-based infrared detector materials such as $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ or CdTe-HgTe superlattices. Growth techniques have been developed to yield high quality heteroepitaxial layers to provide large area, single crystalline CdTe substrates for the subsequent growth of the infrared material. Bulk GaAs has been a popular choice of substrate for the heteroepitaxial growth of CdTe layers of molecular beam epitaxy (MBE). Thick CdTe epilayers on (100) and (111) oriented bulk GaAs have exhibited reasonably good structural and optical properties¹⁻⁵ in spite of the large 14.6% lattice mismatch that is accommodated within a thin highly disordered region at the substrate/epilayer interface.⁶⁻⁸ However, the (100) and (111) orientations still contain numerous macroscopic defects (e.g., hillocks, facets) and point defects that ultimately limit the usefulness of the epilayers in device structures.

Heteroepitaxial growth of CdTe and $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ on the (211)B orientation has received considerable recent interest, due to the ability to suppress twin defect formation along this epitaxial growth direction as well as control macroscopic defects.⁹ The epitaxial growth of CdTe by MBE on (211)B GaAs has been reported to give rise to dual epitaxy under particular substrate preheat conditions,¹⁰⁻¹² with low temperature photoluminescence (PL)¹⁰ and transmission electron microscopy studies¹¹ suggesting the interfacial quality of the (133)CdTe/(211)GaAs epitaxy is superior to that of the (211)CdTe/(211)GaAs epitaxy.

We report a PL investigation of the point defect structure in unintentionally doped (211)B CdTe/(211)B GaAs epitaxial layers grown by MBE at Martin Marietta Electronics Laboratory (Syracuse, NY). The PL spectra ex-

hibit hereto unexplained features in CdTe/GaAs PL that are, in fact, quite pertinent in evaluating layer quality issues. The PL data give evidence for the diffusion of gallium and arsenic atoms from the substrate into substitutional sites in the growing CdTe layer. Edge emission due to Ga_{Cd} and donor-acceptor pair (DAP) recombination related to Ga_{Cd} donors and As_{Te} acceptors was identified. The compensation present in the samples was determined, using PL spectroscopy, by measuring the shift in peak energy of the DAP band with excitation power. In addition, we observe an emission peak at 1.47 eV with weak phonon coupling, which is related to electron-hole recombination around an extended defect with a binding energy of ~ 130 meV.

II. EXPERIMENT

Undoped CdTe epilayers were grown on (211)B GaAs substrates by MBE. The samples were grown using a single CdTe source with substrate temperatures ranging from 230 to 275 °C. All epilayers studied in this report are greater than $3 \mu\text{m}$ in thickness, thus the PL excitation beam probes the film layer only, and not the interfacial region. Defect etch studies were carried out on the epilayers using the etching solution reported by Nakagawa *et al.*¹³ While this etch does not precisely determine the dislocation density, it serves as a relative measure of sample quality. Etch pit densities measured from the (211)B CdTe/GaAs samples included in this study ranged from less than 10^4 to 10^6 cm^{-2} . Transmission electron microscopy (TEM) measurements by N. Otsuka (Purdue University) verified that the samples with the lowest etch pit densities also had relatively low (less than 10^6 cm^{-2}) dislocation densities and that the heteroepitaxial growth relationship was (211)B CdTe on (211)B GaAs. The sample growth conditions and etch pit densities are summarized in Table I.

TABLE I. Summary of growth conditions, etch pit density measurements, and PL amplitudes for (211)B CdTe/(211)B GaAs epilayers. T_S is the substrate growth temperature; epilayer thickness is based on growth rate or Dektak measurements. PL emission amplitudes were measured with an excitation power density of 28 W/cm^2 . The compensation coefficient E_0 was determined from excitation power dependence studies recorded at a sample temperature of 4.7 K .

Sample	Growth temperature ($^{\circ}\text{C}$)	Thickness (μm)	Etch pit densities ($\times 10^4 \text{ cm}^{-2}$)	PL amplitude (arb. units) for emission peak energies			Compensation E_0 (meV)
				1.594 eV	1.51 eV	1.47 eV	
---	270	4.5	33			11	
	270	4.5	80-100		2.7	10	12.2
	230	6.5	120		1.0	1.3	0.29
	275	5.0	0.75		2.9	5.8	2.0
	270	3.8	(not measured)		3.5	7.8	0.91

*Samples with near-edge transmission at 1.592.

To perform the PL measurements, the samples were held above a liquid He bath in a Janis Super-Varitemp dewar. The sample temperature was controlled by flowing helium vapor across the sample surface. The temperature was measured at a position $\sim 3 \text{ cm}$ above the sample and thus represents an upper limit to the actual sample temperature.

The samples were excited using the 514.5 nm output from a Laser Ionics model 552A argon ion laser. The laser beam was modulated and focused onto the sample surface, with excitation power densities ranging from 9 to 310 W/cm^2 . The PL signal was detected using an Instruments SA, Inc. HR-640 spectrometer and a photomultiplier tube with S-1 response along with a lock-in amplifier.

III. RESULTS AND DISCUSSION

A. Edge emission region (1.58–1.60 eV)

Edge PL emission from the (211)B CdTe/(211)B GaAs epilayers at 4.7 K was composed of emission bands at 1.587 and 1.594 eV . Representative emission spectra are shown in Fig. 1 for sample 669A [Fig. 1(a)] and sample 679A [Fig. 1(b)]. An emission band at $\sim 1.594 \text{ eV}$ was observed from all the epilayers included in this study. The

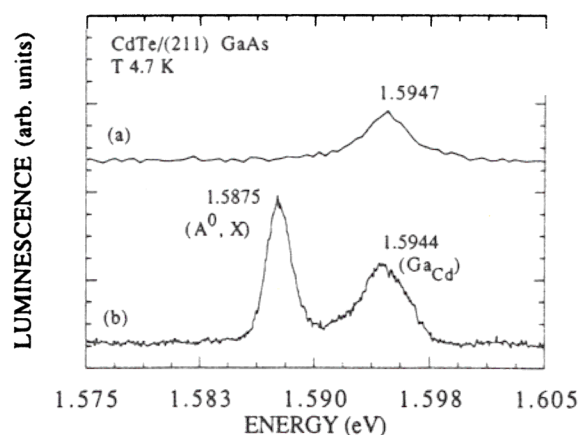


FIG. 1. PL at 4.7 K for (211)B CdTe/(211)B GaAs: (a) sample 669A and (b) sample 679A. Spectra excited using 28 W/cm^2 of 514.5 nm output from argon ion laser.

full width at half maximum (FWHM) of the 1.594 eV emission band was between 3 and 4 meV at liquid helium temperatures. The high energy tail of this emission band extended well above the free exciton recombination energy in CdTe, that occurs at 1.596 eV at 4.7 K , and, as shown later, this band contains a contribution from free exciton recombination.

PL emission at about 1.593 – 1.594 eV in high purity bulk CdTe has been related to recombination of excitons bound to shallow donors such as Ga, In, and Cl that all have ionization energies in CdTe of $\sim 14 \text{ meV}$.^{5,14-16} Such bound exciton recombination, however, is typically composed of sharp emission lines with widths of much less than 1 meV . The intentional introduction of shallow (14 meV) donor species at levels at or above 10^{16} cm^{-3} in CdTe, however, has been shown to produce broad ($> 1 \text{ meV}$) donor bands in edge emission PL spectra for In_{Cd} ^{17,18} and I_{Te} ,¹⁹ where free exciton and bound exciton recombination cannot be separately resolved from the free-to-bound recombination (D^0, h). The substitutional donor Ga_{Cd} is expected to produce a similar broad donor band. Gallium is a relatively fast diffusion species in CdTe, and can originate from the GaAs substrate. Thus we take the presence of the broad emission band at 1.594 eV as evidence of Ga_{Cd} at levels of about 10^{16} cm^{-3} . Indeed, the presence of Ga_{Cd} centers in unintentionally doped layers as thick as $36 \mu\text{m}$ was determined using selective excitation PL spectroscopy on CdTe/(100)GaAs grown by hot-wall epitaxy,⁵ and Ga diffusion in MBE-grown epilayers of CdTe/GaAs, and ZnTe/GaAs has been verified using secondary ion mass spectroscopy.²⁰

Samples 601 and 608C exhibited PL edge emission with similar line shape and width to that shown in Fig. 1(a), however, the peak energy was shifted slightly to 1.592 eV . The shift in energy is believed due to the relative contributions of free exciton and donor-bound exciton recombination, which may be directly related to differences in donor concentrations in the epilayers. The relative amplitudes of the donor edge emission bands at 1.594 eV (or shifted slightly to 1.592 eV) are shown in Table I.

To further characterize the nature of the PL edge emission from the (211)B CdTe/(211)B GaAs epilayers, a study of the excitation power dependence of the 1.594 eV

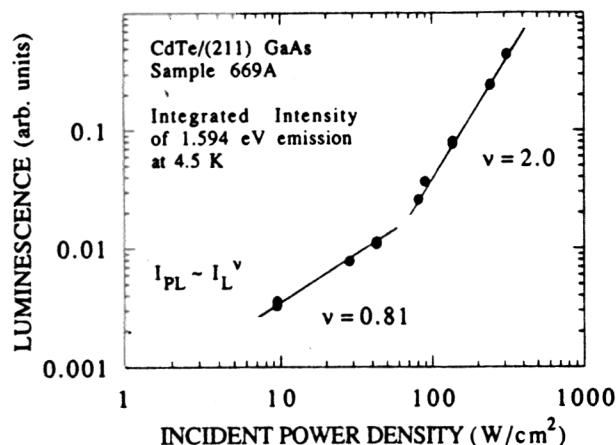


FIG. 2. Integrated near-edge PL emission amplitude of sample 669A as function of incident power density at 4.5 K. Circles are the experimental data. Free exciton recombination is dominant below 60 W/cm².

emission was performed. PL emission intensity generally exhibits the following dependence on incident excitation

$$I_{\text{PL}} = C I_L^\nu, \quad (1)$$

where I_{PL} is the measured PL intensity, C is a constant, I_L is the incident excitation power density (laser power often used instead), and ν is a coefficient whose value is near unity for free exciton emission and significantly greater than unity for bound exciton emissions.^{14,21} The integrated emission intensity for the 1.594 eV band from sample 669 A is shown in Fig. 2 for incident power densities ranging from 9–310 W/cm². Incident power densities above 60 W/cm² gave rise to a superlinear increase in integrated PL intensity with $\nu=2.0$, indicative of bound excitons. For incident excitation below 60 W/cm², the 1.594 eV emission is described by $\nu=0.81$, appropriate for free excitons. The abrupt change in slope directly indicates that the 1.594 eV emission band is indeed a superposition of both free exciton and bound exciton recombination about a shallow donor, presumably Ga. Further, the slight red shift of ~ 1 meV in emission peak energy observed with increasing excitation power density supports the conclusion that bound exciton recombination (i.e., lower emission energy) dominates at higher power densities.

An additional, relatively sharp PL emission at 1.587 eV was measured from samples 679A [Fig. 1(b)] and 694C. The amplitude of this peak quenches rapidly as the sample temperature is raised. At $T > 35$ K, the 1.587 eV emission cannot be observed with an incident power density of 28 W/cm². Such rapid thermal quenching is typical of acceptor-bound exciton (A^0X) recombination. Indeed, (A^0X) emission lines in bulk CdTe in the range 1.588–1.590 eV have been identified as related to several different acceptor impurity atoms, such as Cu, Na, Li, N, P, Ag, and As.²² There is evidence that arsenic from the GaAs substrate has been incorporated into the CdTe epilayers as As_{Te} ; this will be discussed later. However, (A^0X) PL emission related to substitutional arsenic should occur at 1.5897 eV.^{22,23} Because the measured bound exciton emis-

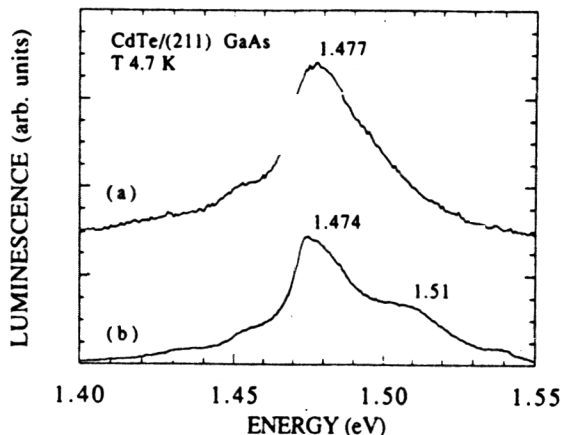


FIG. 3. PL spectra at 4.7 K showing deep level emission bands for (a) sample 601 and (b) sample 608C. DAP band at 1.51 eV is related to Ga_{Cd} and As_{Te} centers.

sion energy from the epilayers is lower than the reported (A^0X) emission energies in bulk CdTe, we believe the defect center related to the 1.587 eV emission is not a simple substitutional site but is likely due to a complex center or extended defect.

B. Deep level emission (1.42–1.55 eV)

At energies well below the edge emission region, broad emission bands were observed from the (211)B CdTe/(211)B GaAs epilayers. Figure 3 shows the deep level emission recorded at 4.7 K, with an excitation of 28 W/cm² for samples 601 [Fig. 3(a)] and 608C [Fig. 3(b)]. The main features are two broad peaks at approximately 1.47 and 1.51 eV, with phonon replicas of the 1.47 eV emission occurring at ~ 1.45 and ~ 1.43 eV. The 1.51 eV band was not resolved in the low temperature PL spectra from sample 601 and appears at most as a shoulder. The relative PL amplitudes of the deep emission bands from the (211)B CdTe/GaAs samples are given in Table I.

A common method of "evaluating" layer quality by the PL technique is to determine the ratio of deep level to near-edge luminescence, the radiative defect density ρ .^{14,24} In the case of relatively high doping levels that can lower radiative efficiency and produce midgap electronic energy levels as competing radiative centers with edge PL centers, it must be pointed out that this method may no longer be a valid procedure for determining epilayer quality. The exact nature or origin of the edge and deep level emission bands must be taken into account when making quality judgments of epilayers used for subsequent deposition. Note from Table I that the relative amplitudes of the edge emission peaks are less than the amplitudes of the deep level emission bands. However, sample 679A was measured by TEM as having a low line dislocation density ($< 10^6$ cm⁻²), appropriate for the subsequent deposition of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ for high performance device applications.

In general, DAP recombination is observed from CdTe in the energy range from 1.4 to 1.55 eV. The DAP band related to a shallow 14 meV donor and the As_{Te} acceptor

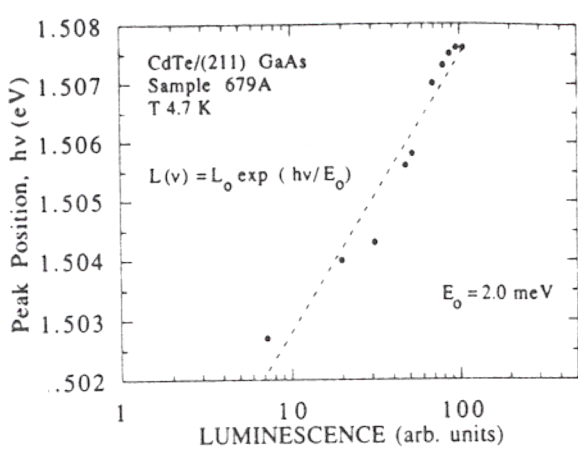


FIG. 4. Energy shift of DAP band at 1.51 eV from (211)B CdTe/(211)B GaAs (sample 679A) with increase in PL emission amplitude, yielding a compensation coefficient $E_0 = 2.0$ meV (dashed line is curve fit).

($E_A = 92$ meV) has been shown to occur at 1.510 eV in bulk CdTe.^{22,23} Thus, the 1.51 eV PL feature indicates that diffusion of As atoms into the CdTe epilayers has occurred in addition to the diffusion of Ga atoms. This evidence for As_{Te} centers is present in the PL spectra for all the films except for sample 601. Therefore, while there is Ga present in sample 601, PL measurements indicate little (if any) As in substitutional sites.

The DAP emission spectrum from a closely compensated semiconductor with approximately equal amounts of donors and acceptors will shift to higher energies as the excitation rate is increased. The peak position E_P depends on the PL emission intensity $L(\nu)$ according to

$$L(\nu) = L_0 \exp(E_P/E_0), \quad (2)$$

where E_0 is the phenomenological compensation coefficient.²⁵ For samples that have moderate or even heavy doping levels, the DAP emission spectrum will not shift significantly. Therefore the amount of compensation in a semiconductor can be determined by monitoring the relative increase in DAP peak energy with increasing PL emission intensity. Samples that have roughly similar concentrations of donors (e.g., Ga) and acceptors (e.g., As) will display large E_0 values.

The measured peak position of the DAP band at 1.51 eV from sample 679A as a function of emission intensity is shown in Fig. 4. Based on power dependence studies, the contribution to the 1.51 eV emission intensity from the closely lying 1.47 eV band is small. Using Eq. (1), the rate of increase in emission intensity with incident excitation power is described by $\nu \sim 0.9$ for the 1.51 eV PL band, and $\nu \sim 0.5$ for the 1.47 eV band.

The shift in emission peak energy shown in Fig. 4 is described by $E_0 = 2.0$ meV. For reference, a shift described by $E_0 = 2.7$ meV was observed for *n*-type GaAs with $N_D - N_A = 3.7 \times 10^{16}$ cm⁻³, and $N_D + N_A = 2.5 \times 10^{18}$ cm⁻³.²⁵ The other (211)B CdTe/(211)B GaAs epilayers included in our study show a range of E_0 values from 0.29 meV (sample 669A) to as high as 12.2 meV (sample

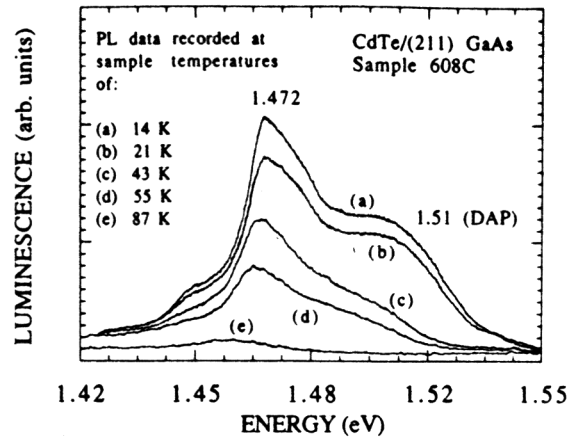


FIG. 5. Temperature dependence of deep level emission from (211)B CdTe/(211)B GaAs (sample 608C). DAP band at 1.51 eV quenches rapidly; 1.47 eV emission still observed at $T > 80$ K.

608C). The compensation coefficients are included in Table I. The observed energy shift of the 1.51 eV emission provides further evidence in support of our identification of the 1.51 eV emission as DAP recombination. It is interesting that the lowest compensation coefficient was measured for the film grown at 230 °C, the lowest substrate temperature investigated in this study. If diffusion of As and Ga is occurring during growth, it would be expected that lower growth temperatures would enhance dissimilarities in As and Ga concentrations.

At an incident power density of 310 W/cm², localized heating may occur at the laser spot on the sample surface, thus increasing the sample temperature and reducing the measured DAP energy shift. However, no increase in sample temperature was indicated by monitoring the energy position of the sharp 1.587 eV emission from sample 679A over the range in power densities used in our study. If sample heating had occurred, the 1.587 eV emission peak would have shifted to lower energies.

The other deep level emission band occurs at 1.47 eV in the CdTe/GaAs epilayers. Although in the spectral region associated with DAP recombination, this feature has a separate origin. The asymmetric line shape of this PL emission [Fig. 3(b)] is caused by the superposition from phonon replicas of the 1.51 eV DAP emission at low temperatures. The emission band at 1.47 eV is too broad to be explained by localized bound exciton recombination. To identify the origin of this recombination in the epilayers, the temperature dependence was recorded.

Deep level PL spectra as a function of temperature from sample 608C are shown in Fig. 5. Note that at 80 K this band is still observed, although shifted slightly to a lower peak energy of 1.468 eV due to the narrowing of the band gap that has occurred with the increase in temperature. PL emission related to shallow donors, such as Ga_{Cd}, is expected to be significantly reduced by thermal quenching at this elevated temperature. For example, the 1.51 eV DAP emission band is no longer observed. Thus the 80 K spectrum indicates that the 1.47 eV emission observed

from (211)B CdTe/(211)B GaAs epilayers is not DAP recombination involving shallow donors.

An additional characteristic of the 1.47 eV emission that also precludes DAP recombination is the weak phonon coupling producing replicas at 1.45 and 1.43 eV. The intensities of phonon-assisted radiative transitions involving LO lattice vibrations, separated by an energy of $E_{LO}=21.3$ meV in CdTe, will follow a Poisson distribution dependence

$$I(h\nu - nE_{LO}) \sim e^{-S} S^n / n!, \quad (3)$$

where $h\nu$ is the zero-phonon transition energy, n is the order of phonon-assisted recombination (one phonon, two phonon, etc.), and S , the Huang-Rhys factor, is the mean number of emitted phonons.²⁶ PL emission between 1.45–1.49 eV in CdTe generally has an $S > 1$ dependence, as has been reported for DAP emission related to Ag_{Te} at 1.492 eV ($S=1$) and Cu_{Cd} at 1.45 eV ($S=2$).²⁶ The 1.47 eV emission band observed from the (211)B CdTe/(211)B GaAs layers, however, displays a much lower value of $S=0.4$, even though it occurs intermediate to these energy bands. Thus, both the temperature dependence PL data and the phonon coupling displayed by the 1.47 eV emission rule out DAP PL recombination.

The 1.47 eV emission band in CdTe epilayers has been attributed in the past to several different recombination processes, including a $Cd_V - D_{Te}$ acceptor complex at about 135 meV above the valence band^{27,28} or the biaxial stress shifted near-edge emission peak from the GaAs substrate in CdTe/GaAs heteroepitaxy (due to the large lattice mismatch).²⁹ The 1.47 eV emission detected in our experiments does not originate from the GaAs substrate since calculations of the penetration depth of 514.5 nm light in CdTe suggest that most (if not all) of the excitation beam is absorbed by the CdTe epilayers within the first 1 μ m of material. In contrast, in an earlier report²⁸ 1.47 eV PL emission was observed in films less than or about 1 μ m, such that the majority of the PL signal originated from the highly disordered interface region between the film and substrate.

To further eliminate consideration of PL emission from the GaAs substrate as being a contributor to the 1.47 eV emission, we performed PL measurements directly on the GaAs substrate for sample 608C. Two emission bands at 1.512 and 1.497 eV were observed at low temperature. The temperature dependence of these GaAs PL emissions did not, however, match that observed for either the 1.51 or 1.47 eV emission bands from the CdTe epilayer, precluding either from originating in the GaAs.

PL emission at ~ 1.47 eV has been observed from CdTe epilayers heteroepitaxially grown on GaAs,^{2,29} InSb,²⁶ and sapphire²⁴ substrates. Emission at this energy has also been reported for homoepitaxially grown CdTe on bulk CdTe substrates.^{27,30} This would indicate that the 1.47 eV emission band is not specific to the growth of CdTe on GaAs, but has origins indicative of a more general phenomenon related to epitaxy.

The probable origin for the 1.47 eV emission in epitaxially grown CdTe films is a structural defect involving dis-

location loops as described by Dean *et al.*,²⁶ similar to the Y center in ZnSe. The proposed defect center responsible for the electron-hole recombination giving rise to 1.47 eV photon emission, such as a small dislocation loop, extends over several lattice sites. Thus the electronic binding potential is uniformly distributed over several lattice sites and is not localized at a single site such as is the case for bound exciton or DAP recombination. The coupling parameter S is proportional to $1/N$, where N is the total number of sites involved with the recombination center. Therefore, the observed phonon coupling strength of the 1.47 eV emission is reduced from its expected value in CdTe (between 1 and 2) to a much smaller value. The value of $S=0.4$ that was found to describe the PL from sample 608C corresponds to a number of lattice sites between $N=3$ to $N=5$. Further, the general line shape of the 1.47 eV PL emission band reported by Leopold *et al.* for CdTe/(100) GaAs² and by Dean *et al.* for CdTe/InSb²⁶ are similar to the emission line shapes observed from the (211)B CdTe/(211)B GaAs epilayers reported here. The weak phonon coupling observed in CdTe/InSb²⁶ was described by $S=0.2$.

The total binding energy of the center responsible for the 1.47 eV emission in the CdTe/GaAs epilayers can be estimated from the peak of the zero-phonon emission band. The binding energy is, thus, found to be $E_g - E_{PL} \approx 130$ meV. The presence of this recombination center can be readily monitored by standard PL spectroscopy. If, indeed, the center is related to small dislocation loops, the PL technique may be the only way to determine its existence since defects involving only a few lattice sites would be too small to be easily seen by electron microscopy techniques.

IV. CONCLUSIONS

PL spectroscopy was performed on (211)B CdTe/(211)B GaAs epilayers to identify point defects and assign origins to features associated with PL from these heteroepitaxial CdTe layers. Edge emission at 1.594 eV is a signature of Ga diffusion in undoped layers and is composed of free exciton recombination and electron-hole recombination about the shallow donor Ga_{Cd} . The relative FWHM of the 1.594 eV emission can be used to estimate the level of doping. DAP recombination at 1.51 eV is related to the diffusion of both gallium and arsenic into substitutional sites in CdTe and is thus a signature of As diffusion into the layers. Analysis of the power dependence of the 1.51 eV PL peak allowed determination of the relative compensation of Ga_{Cd} and As_{Te} centers in the epilayers. The lowest compensation coefficient was obtained in the film grown at the lowest substrate temperature. This indicates that diffusion of one of the dominant impurity species has decreased relative to the other.

A PL emission band at 1.47 eV, characterized by weak phonon coupling, was a dominant recombination process in all the samples studied. The 1.47 eV emission characteristics agree with an earlier report of an extended recombination center related to a structural defect such as a small dislocation loop. The center responsible for the 1.47 eV emission is not believed to be unique to heteroepitaxy of CdTe on GaAs for it has been observed for heteroepitaxial

growth of CdTe on sapphire and InSb as well as homoepitaxial growth on CdTe. As such, it may be common to MBE growth.

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