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# The Fermi level effect in III–V intermixing: The final nail in the coffin?

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We have shown that doping InGaAs/GaAs quantum well materials with  $10^{19}$  Si/cm<sup>3</sup> causes a time and temperature dependent diffusion process, which can be correlated with group III vacancy formation. This process can be modeled and shown to accurately fit other data in the literature. Samples with silicon doping concentrations below this value have no enhanced interdiffusion, in contradiction to the results of the Fermi level model. These results are shown to be comparable to data for AlGaAs/GaAs interdiffusion with doping concentrations between  $5 \times 10^{17}$  cm<sup>-3</sup> and  $10^{18}$  cm<sup>-3</sup>. We have shown that the position of the Fermi level plays no role in III–V intermixing.

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## I. INTRODUCTION

The interdiffusion of semiconductor heterostructures and the role of impurities on the interdiffusion coefficients have received considerable attention since the early 1980s. In particular, the observation by Laidig *et al.*<sup>1</sup> in 1981 that zinc diffusion could completely intermix superlattices at relatively low temperatures started a search for other means of selectively enhancing the interdiffusion of heterostructures.

In 1985 Kawabe *et al.*<sup>2</sup> used secondary ion mass spectroscopy (SIMS) to study the effect of doping on the intermixing of GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As superlattices which were codoped with both silicon and beryllium. Their results showed that for layers with a silicon concentration of  $7 \times 10^{18}$  cm<sup>-3</sup>, if the beryllium concentration was greater than that of the silicon, then there was no observable intermixing of the superlattice after annealing at 750 °C for 2 h. However, without the beryllium being present in concentrations in excess of the silicon concentration the superlattice intermixes completely. In 1987 Mei *et al.*<sup>3</sup> also used SIMS to study the effect of silicon doping, which was incorporated during growth, on the subsequent interdiffusion of GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As superlattices in greater detail. Their results showed a clear dependence of the interdiffusion coefficient on the incorporated silicon concentration.

The following year Tan and Gösele<sup>4</sup> interpreted this data and showed that it appeared to be in agreement with their predictions for the interdiffusion being mediated by triply charged gallium vacancies and thus depending upon the position of the Fermi level. This Fermi level model has become ubiquitous in the literature despite the inability of any other groups to show enhanced interdiffusion of heterostructures at silicon doping levels below  $\sim 10^{18}$  cm<sup>-3</sup>.

Gillin *et al.*,<sup>5</sup> for example, showed in 1993 that for both InGaAs/GaAs and GaAs/AlGaAs structures there was no measurable enhanced interdiffusion of single quantum wells at silicon concentrations of  $10^{17}$  and  $10^{18}$  cm<sup>-3</sup>, although there was a serious degradation of the optical quality of lay-

ers doped with  $10^{19}$  cm<sup>-3</sup> of silicon. In that article they were unable to measure the enhanced interdiffusion with great accuracy but the results indicated an order of magnitude increase in the interdiffusion in the degraded layers. From the photoluminescence spectra they were able to show that there was the creation of a large number of group III vacancies, which were suggested to have been due to the silicon atoms moving from their group III sites to either group V or interstitial positions. In addition, the results showed no variation in the intermixing for *p*-type Be doping up to  $2.5 \times 10^{19}$  cm<sup>-3</sup>.

More recently, Seshadri *et al.*<sup>6</sup> used *p-i-n* and *n-i-p* structures where the Fermi level is varying throughout a region in which three quantum wells were placed. They found only small variations in the diffusion coefficient with the position of the quantum well within their structure and concluded that the Fermi level was playing no role in the interdiffusion of their samples.

In this work we have used both photoluminescence and Rutherford backscattering to study the annealing behavior of highly silicon doped InGaAs/GaAs single quantum well structures.

## II. EXPERIMENTAL METHOD

All the layers used through this work were grown by molecular beam epitaxy (MBE) on (100) oriented GaAs substrates. The quantum well was a 10 nm layer of In<sub>0.2</sub>Ga<sub>0.8</sub>As which had a 100 nm overlayer of GaAs above it. The layers all had flat doping profiles from the surface to a depth of 1 μm, and were doped with silicon to volume concentrations of  $10^{17}$ ,  $10^{18}$ , or  $10^{19}$  Si/cm<sup>3</sup>.

Rutherford backscattering spectrometry (RBS) was performed using 2 MeV He<sup>+</sup> using a surface barrier detector with an overall energy resolution of 12.8 keV and a detector angle of 160°. These experimental conditions were sufficient to allow us to clearly resolve the In signal from the Ga and As surface peaks. Prior to annealing, samples were encapsulated with  $\sim 30$  nm of silicon nitride in a plasma enhanced chemical vapor deposition system and the annealing was per-

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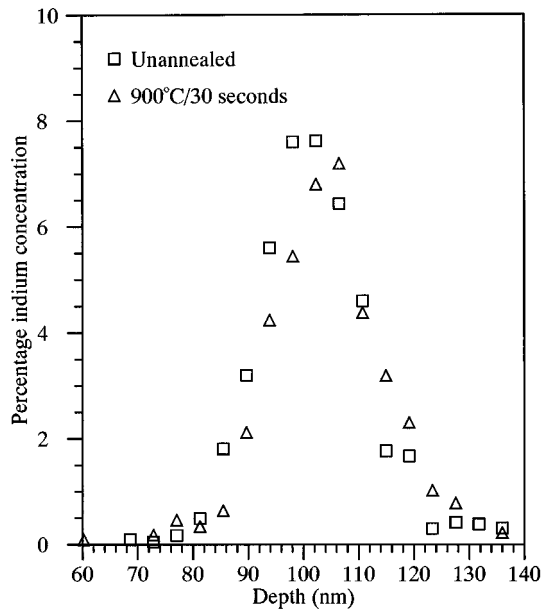


FIG. 1. RBS derived indium profiles, before and after annealing, for a 10 nm  $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$  layer in GaAs doped with  $10^{17} \text{ Si/cm}^3$ .

formed in a double graphite strip heater at temperatures between 700 and 1000 °C in a helium ambient. For all anneals the ramp time from room temperature to the anneal temperature was 15 s. The silicon nitride that we use for our encapsulation has a refractive index of 2.1 and we have characterized our deposition system so that the nitride gives reproducible, low interdiffusion coefficients.

Photoluminescence was excited at a sample temperature of 80 K using the 488 nm line from an argon ion laser at a power density of  $\sim 5 \text{ W/cm}^2$ . The luminescence was dispersed in a 1 m spectrometer and detected with a liquid nitrogen cooled Ge pin diode. All spectra were corrected for the response of the system.

All the samples were measured before annealing, they were then given a 15 s anneal and measured again before receiving a second 15 s anneal and having their final measurements to give the 30 s data.

Details of the theoretical analysis of the photoluminescence spectra used to determine diffusion lengths are given in Ref. 7.

### III. RESULTS AND DISCUSSION

One of the main disadvantages of using RBS to measure the interdiffusion of thin layers is that, unlike photoluminescence, it is not very sensitive to small amounts of diffusion. This is partly due to the fact that the spectra collected are the convolution of the concentration profile with the instrument broadening function, which can be quite large, and partly due to the difficulty in collecting good statistics from thin layers of material. Figure 1, for example, shows the indium concentration profiles derived from RBS spectra for the  $10^{17} \text{ Si/cm}^3$  doped layer before and after annealing at 900 °C for 30 s. From our earlier results using photoluminescence, we would expect this anneal to give a diffusion length of  $\sim 1 \text{ nm}$ . From Fig. 1 very little difference can be seen; however, by repeat-

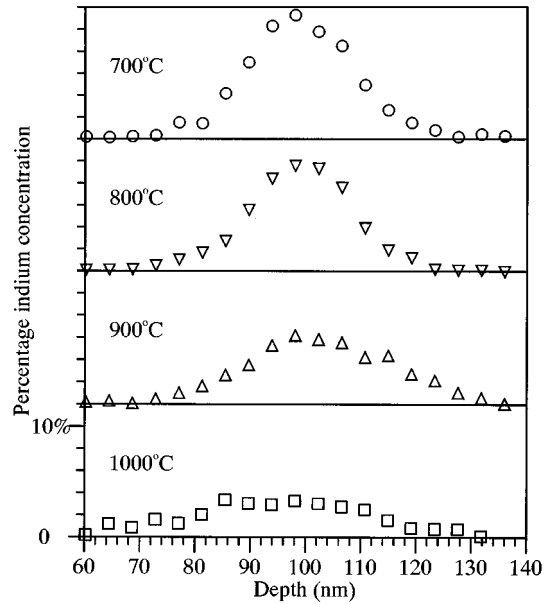


FIG. 2. RBS derived indium profiles, before and after annealing for 15 s at various temperatures, for a 10 nm  $\text{In}_{0.2}\text{Ga}_{0.8}\text{As}$  layer in GaAs doped with  $10^{19} \text{ Si/cm}^3$ .

ing measurements and, using a least squares Gaussian fit to the data, we can calculate the standard deviation for each and show that they all have a sigma of  $10 \text{ nm} \pm 3.5\%$ . This sigma represents the broadening function of the system and any additional broadening due to interdiffusion will be manifest as another Gaussian with a sigma equal to the diffusion length convoluted with this broadening function. As the sigma of two convoluted Gaussians is given by

$$\sigma = \sqrt{\sigma_A^2 + \sigma_B^2} \quad (1)$$

it can be seen that with a repeatability accuracy of 3.5% we will only detect diffusion lengths somewhat greater than 2.7 nm with a similar uncertainty, whereas from photoluminescence data we can determine diffusion lengths of less than 1 nm. From the RBS spectra of the  $10^{18} \text{ Si/cm}^3$  doped sample we were also unable to measure any increase in the sigma of the indium profiles before and after annealing at 900 °C for 30 s. The photoluminescence from both of these samples after annealing was consistent with diffusion lengths of the order of 0.5–1 nm. As we reported in our earlier work,<sup>5</sup> both of these values are comparable to those in undoped material, whereas Mei *et al.*<sup>3</sup> measured an increase in the diffusion coefficient of approximately two orders of magnitude as the doping concentration was increased from  $10^{17} \text{ cm}^{-3}$  to  $10^{18} \text{ cm}^{-3}$ , and this would give an order of magnitude increase in the measured diffusion length, well in excess of our measured sensitivity.

The indium profiles derived from the RBS spectra for the  $10^{19} \text{ Si/cm}^3$  doped layer annealed at various temperatures for 15 s are shown in Fig. 2, and the calculated sigma's are given in Table I. It can be seen that within the experimental sensitivity there has been no interdiffusion of the samples annealed at 700 or 800 °C for 15 s. Figure 3 shows the photoluminescence spectra for the  $10^{19} \text{ Si/cm}^3$  doped sample annealed at 700 °C for 15 and 30 s. From these spectra it can be

TABLE I. The RBS derived values of sigma for the indium profile of the different layers following annealing at various temperatures. Where two values are given these are repeat measurements on the same sample.

Doping concentration	Anneal conditions	RBS calculated sigma (nm)
$10^{17}$ Si/cm <sup>3</sup>	Unannealed	10.44/9.73
$10^{17}$ Si/cm <sup>3</sup>	900 °C/15 s	10.10
$10^{17}$ Si/cm <sup>3</sup>	900 °C/30 s	9.26
$10^{18}$ Si/cm <sup>3</sup>	Unannealed	8.8/9.3
$10^{18}$ Si/cm <sup>3</sup>	900 °C/15 s	9.60
$10^{18}$ Si/cm <sup>3</sup>	900 °C/30 s	9.20
$10^{19}$ Si/cm <sup>3</sup>	700 °C/15 s	9.39
$10^{19}$ Si/cm <sup>3</sup>	700 °C/30 s	9.43
$10^{19}$ Si/cm <sup>3</sup>	800 °C/15 s	9.73
$10^{19}$ Si/cm <sup>3</sup>	800 °C/30 s	11.25
$10^{19}$ Si/cm <sup>3</sup>	900 °C/15 s	12.67
$10^{19}$ Si/cm <sup>3</sup>	900 °C/30 s	13.28
$10^{19}$ Si/cm <sup>3</sup>	1000 °C/15 s	15.95
$10^{19}$ Si/cm <sup>3</sup>	1000 °C/30 s	16.37

seen that the emission from the quantum well is still present, and indicates a diffusion length of  $\sim 1.5$  nm after a 15 s anneal, although a deep level at  $\sim 1.2$  eV is increasing in intensity with annealing. The 1.2 eV level has been attributed to a donor-group III vacancy pair and is evidence that, although there is no catastrophic intermixing, there is the formation of group III vacancies. For the 900 and 1000 °C annealed samples the photoluminescence spectra are very different (see Fig. 4). In both cases the luminescence from the quantum well has completely disappeared and there are broad defect levels at  $\sim 1$  and  $\sim 1.2$  eV. From the sigma values given in Table I we can calculate a diffusion length at 900 and 1000 °C of 7.8 and 12.4 nm, respectively. If these were due to a steady state process they would correspond to diffusion coefficients of  $1.0 \times 10^{-14}$  cm<sup>2</sup>/s and  $2.6 \times 10^{-14}$

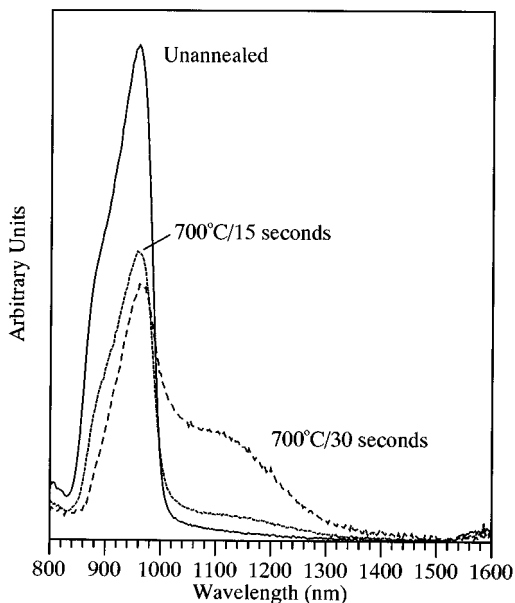


FIG. 3. Photoluminescence spectra for a 10 nm In<sub>0.2</sub>Ga<sub>0.8</sub>As layer in GaAs doped with  $10^{17}$  Si/cm<sup>3</sup>, before and after annealing at 700 °C for 15 and 30 s. The spectra for the two annealed samples have been magnified by a factor of 10.

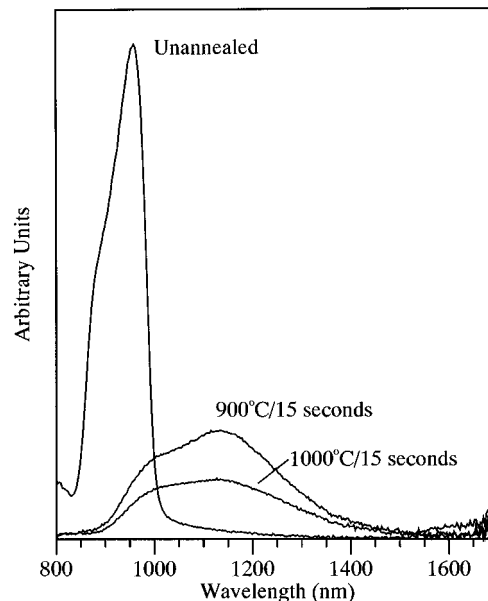


FIG. 4. Photoluminescence spectra for a 10 nm In<sub>0.2</sub>Ga<sub>0.8</sub>As layer in GaAs doped with  $10^{19}$  Si/cm<sup>3</sup>, before and after annealing at 900 and 1000 °C for 15 s.

cm<sup>2</sup>/s, respectively. Using this one can therefore calculate that after a 30 s anneal the diffusion lengths should be 14.8 and 17.7 nm, respectively. These are much greater than the measured values given in Table I. Indeed the sigma's given for the 30 s anneals at 900 and 1000 °C are both within the repeatability error for the measurement, and it would appear that there is relatively little extra mixing during the second anneal. It therefore appears that the rapid intermixing observed at these temperatures is a transient effect and is over within the first 15 s of an anneal.

It is known that silicon acts as an amphoteric dopant and can sit on either lattice site. Studies of silicon ion-implanted GaAs have shown that there is a maximum anneal temperature above which the percentage activation decreases<sup>8</sup> and that this is around 900 °C. If a silicon atom were to move off a group III site (whether it moves to a group V or interstitial site is immaterial), it would leave a group III vacancy behind. Gillin *et al.*<sup>5</sup> have shown from ion implantation experiments that any vacancies on the group III sublattice can diffuse very quickly and cause significant intermixing before being trapped, either at the surface or by other dislocations. Such a process could account for the intermixing behavior observed at these higher anneal temperatures.

It should be noted, however, that even for the samples annealed at 700 °C the defect levels at  $\sim 1.2$  eV are becoming prominent (see Fig. 3). These levels have been associated with a complex between donors and group III vacancies.<sup>9</sup> This would suggest that even at these lower temperatures some of the silicon atoms are moving off group III sites and the vacancies that this creates may be expected to enhance the intermixing if annealed longer. This is indeed seen for the 800 °C annealed samples which, after a 30 s anneal, have measurably diffused.

Most of the data in the literature which show an enhanced interdiffusion in doped materials have been qualita-

tively explained by the Fermi level model. Olmsted *et al.*,<sup>10</sup> for example, measured the interdiffusion of AlGaAs/GaAs superlattices which were grown on undoped or silicon doped substrates ( $n \sim 10^{18} \text{ cm}^{-3}$ ) and were annealed under As-rich and Ga-rich conditions. Under each annealing condition the superlattices showed no dependence of their interdiffusion on the substrate doping, but showed a marked dependence on the annealing ambient. The calculated activation energies for the As-rich and Ga-rich interdiffusion were  $4.91 \pm 0.23$  and  $4.61 \pm 0.21$  eV with  $\ln(D_0)$  values of  $12.70 \pm 2.45$  and  $4.69 \pm 1.22$   $\text{cm}^2/\text{s}$ , respectively. By assuming that the system is in thermodynamic equilibrium they then “normalize” the Ga-rich data to  $P_{\text{As}_4} = 1$  atm and determine a new  $E_A$  and  $\ln(D_0)$  of  $3.26 \pm 0.12$  eV and  $-4.91 \pm 1.22$   $\text{cm}^2/\text{s}$ . This 1.65 eV difference in the activation energies for the As-rich and normalized Ga-rich data is then attributed to a 0.55 eV shift in the Fermi level caused by the generation of  $7 \times 10^{18} \text{ cm}^{-3}$  group III vacancies. Leaving aside the assumptions (most importantly that of thermal equilibrium conditions) used in these calculations, it is interesting to look more closely at the errors in their data. If one takes the data presented in their article and then performs a least squares analysis (ignoring the error bars in both  $D$  and temperature) to determine  $E_A$  and  $\ln(D_0)$  with their statistical errors, the picture is not quite as clear as that presented in the article. For the As-rich conditions we calculated  $E_A$  and  $\ln(D_0)$  to be  $4.8 \pm 0.4$  eV and  $11 \pm 4$ , respectively, while for the Ga-rich conditions we get  $4.6 \pm 0.5$  eV and  $5 \pm 5$ . From this analysis we can see that the activation energies are effectively identical, but with such large errors that any potential differences are well within the experimental error.

The only data which has actually shown a quantitative fit to the theory is that of Mei *et al.*<sup>3</sup> It is therefore necessary to see if the samples used by Mei *et al.*<sup>13</sup> would have had any source of vacancies in them and what effect such a source would have had on their results. The structure used by Mei *et al.*<sup>3</sup> was an AlAsGaAs superlattice in which there was a staircaselike silicon doping profile with concentration plateaus of (I)  $2 \times 10^{17}$ , (II)  $5 \times 10^{17}$ , (III)  $1 \times 10^{18}$ , (IV)  $2 \times 10^{18}$ , (V)  $5 \times 10^{18}$ , and (VI)  $2 \times 10^{18}$ . From this work and from that of Reynolds and Geva<sup>11</sup> it would be expected that layer V would act as a source of vacancies and hence experience a large amount of intermixing.

If layer V is acting as a source of vacancies we can model their subsequent diffusion as a function of time using the standard solution for diffusion of a finite plane

$$N_V(x,t) = N_0 \left[ \text{erf} \left( \frac{h/2 - x}{2\sqrt{D_v t}} \right) + \text{erf} \left( \frac{h/2 + x}{2\sqrt{D_v t}} \right) \right], \quad (2)$$

where  $N_V(x,t)$  is the concentration of vacancies as a function of depth and time,  $N_0$  is the initial concentration of vacancies,  $h$  is the thickness of the initial vacancy layer,  $x$  is the distance in the growth direction (with  $x=0$  at the center of the initial vacancy distribution),  $D_v$  is the diffusion coefficient for the vacancies, and  $t$  is time. As the diffusion coefficient for intermixing is proportional to the vacancy concentration, we can numerically integrate Eq. (2) as a function of time at different depths. Using this we can calculate a term

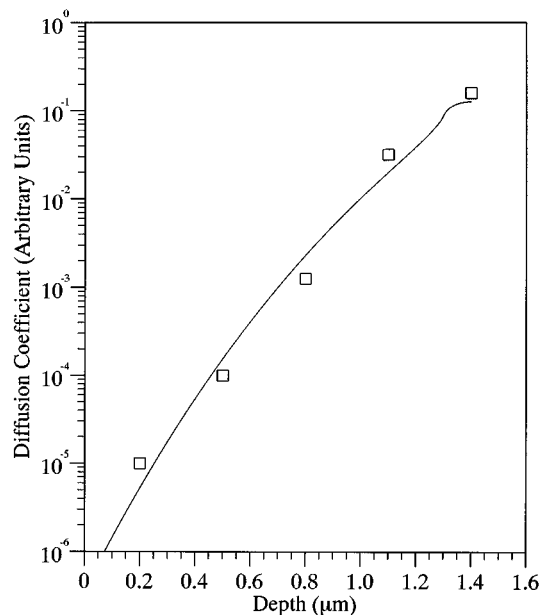


FIG. 5. Diffusion coefficient vs depth derived from Mei *et al.* (Ref. 3) and our fit to the data assuming a source of vacancies centered at a depth of 1.4  $\mu\text{m}$ .

proportional to the effective interdiffusion length for each of the layers in the samples of Mei *et al.* and thus the effective diffusion coefficient they would have measured in their experiment. Figure 5 shows the diffusion coefficient data presented by Mei *et al.* for their 700 °C anneals plotted as a function of depth and the results of our analysis. It can be seen that we get an excellent fit to the data for a diffusion length for the vacancies of 6.8 nm. This corresponds to a diffusion coefficient for the vacancies of  $\sim 6 \times 10^{-16} \text{ cm}^2/\text{s}$  at 700 °C. It should be noted that this diffusion length is the only fitting parameter and the value of  $N_0$  will only move the fit in Fig. 5 vertically. The data points used in Fig. 5 are taken from Ref. 3 where they are presented in arbitrary units. Using our analysis to fit real diffusion coefficients would allow us to calculate both  $D_v$  and  $N_0$ .

Mei *et al.* also observed that the measured intermixing caused by the silicon doping saturates for silicon concentrations greater than  $\sim 10^{19} \text{ Si}/\text{cm}^3$ .<sup>12</sup> Indeed, their data show that the intermixing is inhibited by very high silicon concentrations. As they have shown that these highly doped layers have dislocation loops in them, it is possible that these loops may act as an effective sink for vacancies, removing them before they can cause intermixing. Alternatively, as they gave no electrical data for the silicon doped layers, it is possible that while  $> 10^{19} \text{ Si}/\text{cm}^3$  was incorporated in the layers, not all of these were electrically active (i.e., on group III sites) and hence their subsequent behavior on annealing may not create group III vacancies.

It is now necessary to see if this model can also explain the tellurium doped data collected by Mei *et al.*<sup>13</sup> The first comment to note from the tellurium data of Mei *et al.* is that they measured the samples after both 30 min and 3 h anneals. If the diffusion mechanism in these samples was similar to the one we have described, at a given temperature one would expect to see an apparent diffusion coefficient that would decrease with time. This decrease would be more ap-

parent at higher anneal temperatures where all the vacancies from this process would have diffused evenly throughout the layer during the early stages of the anneal, and this can be seen in their data. It should also be noted that the diffusion coefficient measured for the  $2 \times 10^{18}$  Te/cm<sup>3</sup> layer is two orders of magnitude less than that for silicon doping, and this indicates that there are correspondingly fewer vacancies being created.

Unfortunately, there is much less data available on the annealing behavior and site location of tellurium compared to silicon. However, the fact that tellurium sits preferentially on a group V site makes any mechanisms for the formation of group III vacancies speculative. However, as stated above, the group III vacancy concentrations required to obtain the diffusion coefficients they measured are quite small and one possibility is that at the relatively high doping concentrations used for their study there may have been some tellurium sitting on group III sites which was able to change site with annealing. This suggestion, while speculative, should be compared with the fact that in order for the tellurium data of Mei *et al.* to be fitted to the Fermi level model, Tan and Gösele<sup>14</sup> had to assume that only  $\sim 10\%$  of the tellurium was electrically active.

It is also interesting to speculate on possible causes for the silicon and beryllium data presented by Kawabe *et al.*<sup>2</sup> As stated earlier, they found that superlattice layers doped with either  $6$  or  $7 \times 10^{18}$  Si/cm<sup>3</sup> intermixed completely except where they were co-doped with  $10^{19}$  cm<sup>-3</sup> of beryllium. In their work it can be seen that even in the Be doped layers there is some intermixing near to the interfaces of the Be doped layer as well as deeper into the structure where there is no doping at all. The intermixing in the regions where there is no doping is consistent with the idea of vacancies diffusing out of the doped layer. However, the reduction in intermixing that can be observed with high Be doping may be due to a trapping mechanism for the vacancies. One possible such mechanism would be interstitial beryllium atoms reacting with the vacancies to form an electrically active beryllium acceptors. The use of coimplants of a dopant and a lattice constituent, which sits on the opposite sublattice to enhance electrical activity, has been shown and presumably operates through a similar mechanism.

While these results allow us to explain and quantitatively model the interdiffusion due to highly silicon doped layers, we need to address why these effects are more important than the creation of vacancies by the Fermi level effect and their subsequent effect upon interdiffusion.

Recent work by Khreis *et al.*<sup>15</sup> has used a thin layer of low temperature GaAs as a source of vacancies and used the interdiffusion of a series of quantum wells near the source as a probe for the diffusion of the vacancies. Their work allowed them to measure in a single sample the interdiffusion of InGaAs/GaAs, the diffusion coefficient for vacancy diffusion, and the concentration of point defects present in their samples, all as a function of temperature. The most surprising thing about this work was that they found that the background concentration of vacancies in their material was independent of temperature, at  $\sim 10^{17}$  cm<sup>-3</sup>, between 800 and 1000 °C and was controlled by the concentration of vacan-

cies in the GaAs substrate. This result is in agreement with the work of Dannefaer *et al.*,<sup>16</sup> who determined the vacancy concentration in GaAs wafers to be  $\sim 10^{17}$  cm<sup>-3</sup> at temperatures between 400 and 600 °C.

The work of Khreis *et al.*<sup>15</sup> also shows an Arrhenius plot for InGaAs/GaAs interdiffusion between 750 and 1200 °C. This plot shows no evidence for a higher activation energy at increasing temperatures which one would expect if the thermal equilibrium concentration was becoming higher than the grown in concentration. These results show that, contrary to popular belief, the interdiffusion of III–V materials grown on GaAs substrates is not controlled by a thermal equilibrium vacancy concentration but rather by a much larger concentration of vacancies which are frozen into the substrate during growth.

This result gives us the reason why we would not expect to see changes in the interdiffusion coefficient due to the equilibrium vacancy concentration changes predicted by the Fermi level model.

Thus if III–V materials grown off GaAs substrates have nonequilibrium vacancy concentrations, what about materials grown off InP substrates? Measurements of group III interdiffusion in InGaAs/InGaAs layers grown on InP substrates<sup>17</sup> give an activation energy for interdiffusion which is identical, within experimental error, to that for InGaAs/GaAs grown on GaAs. However, the prefactor for the InP based material is over an order of magnitude greater than that for the GaAs based material. As the work of Khreis *et al.*<sup>15</sup> showed that the activation energy for interdiffusion in InGaAs/GaAs is the activation energy for vacancy diffusion, then the differences in prefactors between interdiffusion on materials grown on InP and GaAs substrates is the difference in the grown in vacancy concentrations.

Therefore InP also has a nonequilibrium vacancy concentration and hence the creation of vacancies by the Fermi level effect in materials grown on InP substrate would again be too small to affect the interdiffusion.

#### IV. CONCLUSION

In conclusion, we have shown that doping InGaAs/GaAs quantum wells with  $10^{19}$  Si/cm<sup>3</sup> causes a temperature and time-dependent fast-intermixing process which can be correlated with the formation of group III vacancies. The effect of the formation of such vacancies in the samples of Mei *et al.*<sup>3</sup> on the subsequent interdiffusion in their samples has been modeled and is shown to give the variation in diffusion coefficient with depth that they observed without the use of a Fermi level model. These data have been the main result on which the Fermi level effect has been based. Experiments on layers with  $10^{17}$  Si/cm<sup>3</sup> and  $10^{18}$  Si/cm<sup>3</sup> doped layers show no enhanced interdiffusion in contradiction to the predictions of the Fermi level model.

Recent results show that the reason for this is that interdiffusion in III–V materials is not governed by thermal equilibrium vacancy concentrations but rather by vacancy concentrations which are grown in to the substrate materials. These results show that the Fermi level would not be expected to play a role in the interdiffusion of real samples as

the Fermi level model assumes that the vacancy concentrations are in thermal equilibrium.

## ACKNOWLEDGMENTS

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