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Analysis of the shallow and deep center occupancies in Si-doped $AI_x Ga_{1-x} As$ using a multilevel donor model

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The concentration of occupied deep centers in Si-doped Al_xGa_{1-x}As for $x \ge 0.2$ has been calculated from a three-level donor model, in which the shallow levels are treated as excited states of the deep (DX) ground state. The deep level is assumed to be tied to the L valley, and the shallow levels to the Γ and X valleys. The behavior of the free-electron density and the thermal activation energy as function of composition is in good agreement with experimental results reported in the literature. In this model of dependent donor levels the deep-level occupancy can be directly calculated without needing deep-level transient spectroscopy measurements. A two-level donor model is used to calculate the pressure dependence of the deep level from a hydrostatic pressure experiment on a GaAs/Al_{0.3}Ga_{0.7}As heterostructure reported in the literature. We assume a shallow level tied to the Γ valley and an arbitrary deep level which is not coupled to any of the conduction bands. The calculation of the position of the deep level relative to the Γ valley as a function of pressure confirms the coupling of the deep level to the L valley. In this dependent donor model no large compensation is needed to fit the experimental data.

I. INTRODUCTION

The electronic properties of $Al_xGa_{1-x}As$ are controlled by the coexistence of a hydrogenlike shallow center and a deep center, the so-called DX center. This deep center, which is dominant for 0.25 < x < 0.6, is also responsible for the effect of persistent photoconductivity (PPC). The nature of the deep center is a long-standing problem. Lang, Logan, and Jaros¹ proposed a large lattice relaxation (LLR) model, whereas Saxena² used a band structure model to explain the features of the deep center. Hydrostatic pressure experiments by Tachikawa et al.³ and Lifshitz, Jayaraman, and Logan⁴ showed that the deep center can be induced by pressure. From these experiments it has been proposed that the deep center is tied to the L minimum and that the predominance of either shallow or deep centers depends on the relative positions of their energy levels. The total number of deep and shallow centers has proven to be nearly equal to the amount of Si doping (Watanabe et al.⁵). Therefore, it has been concluded that both centers are induced by the same donor. Li et al.6,7 showed that the photoionization cross section and the thermal capture and emission energies are the same for the pressure-induced deep centers in GaAs as for the deep centers in $Al_xGa_{1-x}As$. From photoionization measurements it has recently been suggested by Henning and Ansems⁸ that the deep level does not show LLR but only small lattice relaxation (SLR). The same idea has been put forward by Hialmarson and Drummond.9 This has also been supported by EXAFS experiments by Mizuta and Kitano.¹⁰ FIR measurements by Theis et al.¹¹ proved that all the deep centers in Al_{0.4}Ga_{0.6}As act as shallow centers after photoionization. The maximum attainable electron density in highly doped GaAs should be due to self-compensation. At high doping concentrations, however, the conduction band is filled to such an amount that the deep center, which lies above the conduction-band edge, becomes populated.^{12,13} Theis et al.¹⁴ also proved that in GaAs a metastable level is present which can be persistently populated by hot electrons. This level is believed to be the deep center which becomes resonant with the Γ band for x < 0.22.

These experiments strongly suggest that the shallow donor states can be regarded as excited states of the deep (DX)ground states. Assuming such a donor model implies the use of the statistics of a multilevel donor. Saxena¹⁵ already used a two-level donor model to determine the pressure dependence of the band structure. However, he did not point out the consequences of the use of a multilevel donor for the occupancies of the different levels. In the present paper we show that the shallow and deep center occupancies and the freeelectron concentration in Si-doped Al, Ga1_, As as function of the composition can consistently be described on the basis of a dependent three-level donor model, properly taking into account the distribution function. This model proves to be also suitable for analyzing the behavior of an AlGaAs/GaAs heterostructure under hydrostatic pressure.

II. THE MULTILEVEL DONOR MODEL

In previous calculations,^{3,16,17} the occupancies of the shallow and deep centers are given by

$$n_{DX} = \frac{N_{DX}}{1 + g_{DX}^{-1} \exp\left[(E_{DX} - E_F)/kT\right]},$$
 (1)

$$n_s = \frac{N_s}{1 + g_s^{-1} \exp\left[(E_s - E_F)/kT\right]},$$
 (2)

where N_{DX} and N_s are the concentrations of deep and shallow centers, respectively; E_{DX} and E_s are their ionization energies, and g_{DX} and g_s are the degeneracy factors. The shallow and deep centers are then treated as two indepen-

4269

J. Appl. Phys. 66 (9), 1 November 1989

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© 1989 American Institute of Physics 4269 dent kinds of donors. Tachikawa *et al.*³ demonstrated that the electron occupancy of the deep centers can be calculated in a three-level (independent) donor model, assuming that the deep center is coupled to the *L* minimum with $E_L - E_{DL} = 120$ meV. If, however, a donor is assumed to have more than one level, the total donor occupancy cannot be calculated by assuming independent donors. In a situation where the deep centers are dominant, an electron excited from the deep ground state can be trapped at an excited (shallow) state in a dependent donor model. Assuming independent donors, where the dominance of deep centers means that N_{DX} equals the total donor concentration, no trapping at shallow centers is possible because these shallow centers are transferred into deep centers. So the use of (1) and (2) is not correct as was also pointed out by Morgan.¹⁸

In the work presented here we show that the occupancy of the deep center and the free electron concentration can be derived by assuming a donor with one ground state and two excited states. The ionized donor concentration is then given by (Cohen¹⁹)

$$N_{D}^{+} = \frac{N_{D}}{1 + \sum_{r} \{g_{r} \exp[(E_{F} - E_{dr})/kT]\}}, \quad r = \Gamma, L, X.$$
(3)

Using Eq. (1) the deep center concentration N_{DX} has to be determined for various compositions by deep-level transient spectroscopy (DLTS) measurements. In our calculation, using Eq. (3), deep center occupancies can be calculated without knowing any experimental data concerning the deep centers.

Using a multivalley effective-mass model, Chand et al.20 calculated the activation energy when there are different energy levels connected to the Γ , L, and X bands. Watanabe and Maeda¹⁷ suggested that the variation in activation energy of the donor as a function of composition is due to a variation in the concentration ratio between shallow and deep centers. In the present paper we show that the variation in the activation energy is due to different occupancies of the ground and excited states of the donors. From this model of dependent donor levels, it is also easily understood why the deep center activation energy for emission is independent of the composition x (Watanabe *et al.*⁵), while the activation energy for capture decreases with increasing x (Mooney, Caswell, and Wright²¹). For x > 0.22 the shallow state becomes the excited state of the deep ground state. After photoionization of an occupied deep state, the electron will only return to this deep state if the thermal energy is large enough to overcome the potential barrier between the ionized and bound state.

In our calculations, we assume a shallow level tied to the Γ minimum, a deep level tied to the *L* minimum, and a shallow level tied to the *X* minimum. For the constant energy separation $E_L - E_{DL}$ between the *L* minimum and the coupled deep center, we use a value of 160 meV. From Hall and DLTS measurements, values of 120 and 140 meV had been found by Tachikawa *et al.*³ and Lifshitz and co-workers,⁴ respectively. From photoluminescence measurements Henning and Ansems⁸ found that $E_L - E_{DL} = 200$ meV, and Calleja, Gomez, and Munoz²² reported 220 meV; so there

seems to be a large discrepancy between the various values reported. Comparing these results, one has to keep in mind that the deep centers start to dominate the conduction for x > 0.2 because at this composition the deep centers cross the Γ band. The different values of $E_L - E_{DL}$ are partly due to the differences in the band-structure models used by several authors. This is shown in Table Ia where for several models $E_L - E_{\Gamma}$ at x = 0.2 is given. Taking into account the differences in the several band-structure models, the discrepancy in $E_L - E_{DL}$ of about 80 meV reduces to 40 meV in $E_{\Gamma} - E_{DL}$. In our calculations the band-structure model reported by Casey and Panish²³ is adopted. In this band-structure model, the activation energy of the deep center at x = 0.3 is about 60 meV, a value commonly accepted in cal-GaAs/Al_{0.3}Ga_{0.7}As heterostructures. culations on Ishikawa et al.²⁴ also reported an ionization energy of 60 meV at x = 0.3. For g_r and E_{dr} we use the values as given in Table Ib. The temperature dependence of the band structure is taken into account by using the Varshni²⁵ equation

$$E_g(T) = E_g(300) + \alpha_r(300)^2 / (504) - \alpha_r T^2 / (T + \beta_r),$$
(4)

with coefficients α , and β , given by Aspnes, ²⁶

$$\alpha_{\Gamma} = 5.41 \times 10^{-4} \text{ eV } \text{K}^{-1},$$

$$\alpha_{L} = 6.05 \times 10^{-4} \text{ eV } \text{K}^{-1},$$

$$\alpha_{X} = 4.60 \times 10^{-4} \text{ eV } \text{K}^{-1},$$

$$\beta_{\Gamma,L,X} = 204 \text{ K}.$$

The electron density in each conduction band, using the Ehrenberg approximation (Blakemore²⁷), is given by

$$n_r = \frac{N_{cr}}{\frac{1}{4} + \exp[(E_{cr} - E_F)/kT]}, \quad r = \Gamma, L, X, \quad (5)$$

with

$$N_{cr} = 2(2\pi m_r^* kT/h^2)^{3/2}$$

For m_r^* the values reported by Joyce²⁸ are used. Neglecting the acceptor concentration, charge neutrality requires

$$n_{\Gamma} + n_{L} + n_{X} = N_{D}^{+}.$$
 (6)

After obtaining E_F from Eqs. (3), (5), and (6) for different AlAs fractions, the occupancy of each state can be calculated by (Cohen¹⁹)

$$n_{dr} = N_D^+ g_r \exp\left(\frac{E_F - E_{dr}}{kT}\right).$$
⁽⁷⁾

The magnitude of the doping concentration N_D does not influence the relative occupancy of the donor levels.

III. ANALYSIS OF EXPERIMENTAL DATA

A. Al, Ga1_, As

Figure 1 presents the results of our calculations of the occupancies of the donor levels and the free-electron concentration at room temperature. For the doping concentration a value of $N_D = 1 \times 10^{23} \,\mathrm{m}^{-3}$ is used. The deep center concentration (DLTS), the shallow donor concentration (C-V), and the free-electron concentration (Hall measurements) at $T = 300 \,\mathrm{K}$ reported by Watanabe *et al.*⁵ are also given. The shape of the calculated free-electron concentration agrees

4270 J. Appl. Phys., Vol. 66, No. 9, 1 November 1989

Іа	Band str. model	$E_L - E_{\Gamma} (x = 0.2)$ (meV)	$\frac{E_L - E_{DL}}{(\text{meV})}$	$E_{\Gamma} - E_{DL} (x = 0.2)$ (meV) -43 22			
Tachikawa <i>et al.</i> ^a	Casey and Panish ^b	163	120				
Henning and Ansems ^d	Saxena $(T=0)^e$	200	200	- 23			
Present paper	Casey and Panish ^o	163	160	- 3			
ІЬ		$T = 300 \text{ K}^{\text{b}}$					
$E_{d\Gamma} = E_{\Gamma} - 6 \text{ meV}^{a}$	$g_{\Gamma}=2$	$E_{c\Gamma}(x) = 1.424 + 1.247x, x < 0.45 (eV)$ = 1.42 + 1.247x + 1.147(x - 0.45) ² , x > 0.45					
$E_{dL} = E_L - 160 \text{ meV}$	$g_L = 8$	$E_{cL}(x) = 1.708 + 0.642$	x(eV)				
$E_{dx} = E_x - 40 \text{ meV}^a$	$g_{\chi} = 6$	$E_{cX}(x) = 1.900 + 0.125x + 0.143x^{2}(eV)$					

^a M. Tachikawa, M. Mizuta, H. Kukimoto, and S. Minomura, Jpn. J. Appl. Phys. 24, L821 (1985).

^b H. C. Casey and B. Panish, *Heterostructure Lasers* (Academic, New York, 1978).

^eN. Lifshitz, A. Jayaraman, and R. A. Logan, Phys. Rev. B 21, 670 (1980).

^d J. C. M. Henning and J. P. M. Ansems, Semicond. Sci. Technol. 2, 1 (1987).

^eJ. C. M. Henning, J. P. M. Ansems, and A. G. M. de Nijs, J. Phys. C 17, L915 (1984).

rather well with the experimental data. For low values of xthe free-electron concentration decreases due to an increasing occupancy of the deep center with increasing x. For x > 0.45 the lowest conduction band changes from Γ to X, and the high effective density of states of the X band gives rise to an increasing free-electron concentration. We find the minimum in the electron concentration at about x = 0.42. Ishibashi, Tarucha, and Okamoto²⁹ and Watanabe et al.⁵ reported experimental values of x = 0.36 and 0.45 for this minimum. A precise determination of this minimum is difficult because measurements on different samples have to be compared. The results obtained at T = 100 K are plotted in Fig. 2 together with the DLTS data reported by Lang and coworkers.¹ As expected, the donor with an occupied deep ground state is dominant for 0.25 < x < 0.6. It should be noted, however, that the positions of the DLTS peaks, from which the occupancy has been determined, lie at different temperatures for different x. So the measured and calculated values (at one temperature) are not really comparable. Taking this into account, the calculated occupancy of the deep centers is in good agreement with the DLTS data. At T = 100 K the free-electron concentration is also reduced by the high occupancies of the shallow centers.

In order to determine the activation energy, the temperature dependence of the free-electron concentration has been calculated for several values of x in the temperature range between 100 and 300 K. For temperatures lower than 100 K no thermal equilibrium can be maintained; the electrons cannot be trapped in the deep centers anymore because of the potential barrier between the bound and ionized state. When Boltzmann statistics are applicable, the condition of charge neutrality gives (Blakemore²⁷)





FIG. 1. Occupancy of the donor levels and the free-electron concentration at room temperature as a function of AlAs mole fraction x.

4271 J. Appl. Phys., Vol. 66, No. 9, 1 November 1989

FIG. 2. Occupancy of the donor levels and free-electron concentration at T = 100 K as a function of AlAs mole fraction x in comparison with the DLTS data reported by Lang and co-workers (see Ref. 1).

Blom et al. 4271

 $g_r(N_A + n)n/(N_D - N_A)N_c = \exp(-E_D/kT).$ (8)

Because in our calculation zero compensation is assumed, the thermal activation energy ΔE can be defined by the relation

$$n \simeq \exp(-\Delta E/2kT).$$

The calculated results for the activation energy ΔE are shown in Fig. 3. This curve agrees very well with the curves determined from Hall measurements by Ishibashi and co-workers,²⁹ Ishikawa *et al.*,²⁴ and Chand *et al.*²⁰

B. Al_xGa_{1-x}As/GaAs heterostructure under hydrostatic pressure

The multilevel donor model is also useful in describing the properties of a GaAs/Al_xGa_{1-x}As heterostructure. From hydrostatic pressure measurements up to 16 kbar³⁰ on a GaAs/Al_{0.3}Ga_{0.7}As heterostructure, the pressure dependence of the deep center can be determined. An advantage of pressure measurements is that only one sample is needed to study the changes in ionization energy of the deep center. Mercy *et al.*³⁰ neglected the free-electron concentration in the *L* and *X* bands. At a pressure of about 15 kbar, however, the conduction-band minimum changes from Γ to *X*; so for this pressure the free-electron concentration in the *L* and *X* bands cannot be neglected. We assume a shallow level tied to the Γ band and an arbitrary deep level with degeneracy factor g_{dl} . The parameters used are defined in Fig. 4 and Table



FIG. 3. Thermal activation energy as a function of AlAs mole fraction.

II. The position of this deep level is now defined relative to the bottom of the well by an energy E_{dl} ; so no coupling between the deep level and one of the conduction bands is assumed. The pressure dependence of the deep level can be determined by calculating the positions of the deep level (E_{dl}) and the Γ valley (E_c) , both relative to the bottom of the well, as a function of pressure.

In the AlGaAs side (z>0) we have the Poisson equation $\nabla^2 v = q\rho(z)/\epsilon_0\epsilon_r$. The charge density is given by $\rho(z) = q[N_d^+(z) - N_A - n(z)]$, where

$$N_{d}^{+}(z) = \frac{N_{d}}{1 + 2 \exp\left\{\left[E_{F} - E_{sl} - v(z)\right]/kT\right\} + g_{d1} \exp\left\{\left[E_{F} - E_{d1} - v(z)\right]/kT\right\}},$$
(9)

$$n_r(z) = \frac{N_{cr}}{\exp\{[E_{cr} - E_F + v(z)]/kT\} + (1/4)}, \quad r = \Gamma, L, X,$$

$$n(z) = \sum_r n_r(z).$$
(10)

Integration of the Poisson equation gives

$$-\left(\frac{dv}{dz}\right)^{2}\Big|_{z=0} = \frac{2kTN_{D}q^{2}}{\epsilon_{0}\epsilon_{r}} \left[\ln\left(\frac{2\exp\left[\left(E_{F}-E_{sl}\right)/kT\right]+g_{dl}\exp\left[\left(E_{F}-E_{dl}\right)/kT\right]+1}{2\exp\left[\left(E_{F}-E_{sl}\right)/kT\right]+g_{dl}\exp\left[\left(E_{F}-E_{dl}\right)/kT\right]+\exp(V_{0}/kT)}\right) + \frac{N_{A}V_{0}}{kT} + 4\Sigma_{r}N_{cr}'\ln\left(\frac{\exp\left[\left(E_{cr}-E_{F}\right)/kT\right]+1/4}{\exp\left[\left(E_{cr}-E_{F}\right)/kT\right]+1/4\exp\left(-V_{0}/kT\right)}\right)\right],$$
(11)
$$N_{cr}' = N_{cr}/N_{D}.$$

Far from the junction, where v(z) = 0, charge neutrality requires

$$n(z) + N_{A} = N_{D}^{+}(z).$$
(12)

The electrical field remains constant over the spacer; so Eq. (11) can be rewritten using

$$\left. \frac{dv}{dz} \right|_{z=0} = \frac{q^2(n_{2\deg} + n_{\deg})}{\epsilon_0 \epsilon_r}$$

where n_{depl} is the concentration of the depletion charge on the GaAs side. From the energy-band diagram, we have

$$\Delta E_c = E_F + \delta + V_0 + \frac{q^2 (n_{2\text{deg}} + n_{\text{depl}})}{\epsilon_0 \epsilon_r} d_1. \quad (13)$$

The Fermi energy E_F can be determined from n_{2deg} by using the triangular well approach (Stern³¹). The energy level E_0 is then given by

$$E_0 = (\hbar^2/2m^*)^{1/3} \left[\frac{3}{2} \pi e \langle F \rangle (i + \frac{3}{4}) \right]^{2/3}, \tag{14}$$

where $\langle F \rangle$ is the average electric field felt by the electrons in the well, defined as

$$\langle F \rangle = \frac{e(n_{\rm depl} + 0.5n_{\rm 2deg})}{\epsilon_0 \epsilon_r} \,. \tag{15}$$

If we assume that $E_{\rm sl} = 6 \text{ meV}$ (the shallow level plays no

Blom et al. 4272

4272 J. Appl. Phys., Vol. 66, No. 9, 1 November 1989



FIG. 4. Band-structure diagram of an Al_xGa_{1-x}As-GaAs heterostructure, for x = 0.3, $N_D = 7.5 \times 10^{23}$, and $d_1 = 60$ Å.

important role at x = 0.3), we have three equations, (11), (12), and (13), to determine the three unknown parameters $E_{\rm dl}$, V_0 , and δ . When $n_{\rm 2deg}$ and the compensation ratio are known, the position of the deep center in the AlGaAs region can be calculated. Because we do not know to which conduction band the deep level is coupled, the calculation has to be done for the three possible values of $g_{\rm dl}$ (see Table I). By using the pressure dependence of $n_{\rm 2deg}$ at room temperature, as measured by Mercy *et al.*,³⁰ we calculated the pressure dependence of ratio are compensation ratio at the pressure dependence of ratio at the pressure dependence of the deep center for several compensation ratio at the pressure dependence of ratio at the pressure dependence of ratio at the pressure dependence of the deep center for several compensation ratio at the pressure dependence of the deep center for several compensation ratio at the pressure dependence of the deep center for several compensation ratio at the pressure dependence of the pressure

TABLE II. Parameters of $Al_xGa_{i-x}As$ used in the calculations and calculated results for the position and the pressure dependence of the deep center.

Conduction-band discontinuity $\Delta E_c = 0.65 \times 1$.	$247 \times 0.3 = 243 \text{ (meV)}$
Pressure coefficient of E_{cr}^{a}	(meV/kbar)
dE_{eV}/dp	11.11
dE_{cL}/dp	2.8
dE_{cx}/dp	- 0.8

Calculated results

 $g_{dl} = 2$

$\frac{E_c - E_{dl}(p=0)}{(\text{meV})}$	$d(E_c - E_{dl})/dp$ (meV/kbar)		
95.2	8.8		
82.7	8.7		
69.3	8.8		
66.8	8.8		
54.3	8.7		
41.4	8.8		
59.4	8.8		
46.8	8.7		
33.6	8.8		
	$E_{c} - E_{dl}(p = 0)$ (meV) 95.2 82.7 69.3 66.8 54.3 41.4 59.4 46.8 33.6		

^aS. Adachi, J. Appl. Phys. 58, R1 (1985).

4273	J. Appl.	Phys.,	Vol.	66,	No.	9,	1	November	1989
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tios. The results are given in Table II. Mercy *et al.*³⁰ have chosen $N_A/N_D = 0.32$ and $d(E_c - E_{\rm dl})/dp = 11$ meV/kbar to fit the experimental values. In our calculation no large compensation is needed to fit the experimental data. If coupled to the *L* minimum ($g_{\rm dl} = 8$), we find for $N_A = 0$ at zero pressure that $E_c - E_{\rm dl} = 59.4$ meV. In this case the energy separation between the *L* minimum and the deep center turns out to be 161.9 meV, which agrees very well with the value assumed previously in our analysis.

According to Adachi³² the differences between the pressure coefficients of the Γ and L bands and the Γ and X bands are 8.3 and 11.9 meV/kbar, respectively. The calculated pressure dependence of the deep center, independent of the choice for $g_{\rm dl}$, once more confirms the coupling of the deep center to the L band.

IV. CONCLUSIONS

In conclusion, we showed that the use of a multilevel deep donor model gives an adequate description of the general features of the deep center occupancy and the free-electron concentration in bulk AlGaAs. Using this model, no DLTS measurements are needed to determine the deep center concentration. Applying this model to a GaAs/AlGaAs heterostructure under hydrostatic pressure, confirms the idea that the deep center is coupled to the L minimum with a binding energy of about 160 meV.

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Blom et al. 4273

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4274 J. Appl. Phys., Vol. 66, No. 9, 1 November 1989