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The utilization of DX centres in high-pressure studies of low-dimensional doping structures in GaAs

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Abstract. High-pressure experiments with thin slabs of silicon donors in GaAs are employed to search for the correlation effects expected from the formation of D^+D^- pairs expected from the negative-U model of DX centres. The mobilities in the individual subbands are very dependent on subband energy but a preliminary analysis does not require the existence of such pairs.

1. Introduction

The ability of DX centres to trap out conduction electrons metastably can be exploited in quantum transport experiments with GaAs structures containing planes of silicon donors introduced during MBE growth. The results from earlier work involving the Imperial College group with such delta wells and thin doping slabs [1] are reviewed and discussed with respect to the different models for DX centres.

Transport measurements with bulk-doped samples by Maude *et al* [2] showed a large increase in mobility on increasing the pressure due to the change in charge state of the localized states. Controversy has arisen concerning the interpretation of this change in mobility. It was initially argued that the sign of the pressure-induced change in mobility provided conclusive qualitative evidence against the negative-U model of DX centres [2, 3]. However, it was shown later that a mobility increase of the required sign and magnitude could result from impurity correlation effects [4, 5]. Experiments with delta-doped samples provide an additional way to investigate this possibility as the extent of the electronic wavefunctions in the z-direction increases rapidly with increasing subband energy. Consequently, if the dipolar fields expected from correlation effects are present, the mobility changes should be greater for the higher-order subbands than for the $i = 0$ case (which should 'see' only monopolar fields). Furthermore the spacing of the individual subbands should be modified by the dipolar component of the potential local to the doping plane. The electrons are expected to trap out onto donor sites which are favoured energetically and thus have an ionized (positively charged) donor close by. On a negative-U model, D^+D^- pairs will form and the dipolar component will be very strong. On the positive-U picture there will be no negative impurities present unless compensating acceptors are introduced during growth. Consequently the dipolar component of the local potential will be weak. The results of reference [1] are analysed in an attempt to detect correlation effects.

In reference [1] a method was developed for determining the individual subband mobilities quantitatively from the width of the peaks resulting from the Fourier analysis of the Shubnikov–de Haas oscillations. It was found that the mobility in the individual subbands can differ by as much as an order of magnitude but all the subband mobilities increase with increasing pressure by about the same amount. Consequently our initial interpretation appears to confirm the conclusions of Maude *et al* [2, 3] concerning the change in charge state of the Si donors. Another important result to emerge from this study is that Hall mobility measurements for delta samples must be analysed using a full multicarrier model involving estimates of the individual subband model. The Hall mobility as measured with the thinnest of the current samples can actually fall with increasing pressure, although all mobilities within the individual subbands are increasing. These apparently conflicting results can be reconciled on a multicarrier model, with the fall in the Hall mobility simply arising from the depopulation of higher-mobility upper subbands which are weighted more heavily in Hall experiments.

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2. Experimental techniques

Three GaAs samples with thin doping Si slabs were grown by MBE at Philips Research Laboratories at Redhill. The growth temperature was 400 °C. The value for the areal concentration was constant at a value of 1.1×10^{13} cm^{-2} and the slab thicknesses assuming no spreading of the Si donors were 2.0, 5.0 and 10 nm giving volume concentrations of 5.5×10^{19}, 2.2×10^{19} and 1.1×10^{19} cm^{-3} respectively. The thickness of the epilayer was 1 μm in each case and the doping slab was located 0.5 μm from the surface. The doping slabs were formed by sequential planar deposition of silicon at the appropriate doping level; e.g. the 5 nm slab was formed by depositing 18 planes of 5.56×10^{11} cm^{-2} of silicon separated by 2.8 Å. Local vibrational mode (LVM) studies of single-delta wells grown at 400 °C in the same reactor and having an areal concentration of 2×10^{13} cm^{-2} did not show any sign of silicon switching to the As site or of the formation of Si_{Ga}–Si_{As} pairs above the detection limit [6]. The volume concentration in these samples (estimated on the assumption that no spreading had taken place) would have been more than an order of magnitude higher than the concentrations employed in the present work. The temperature of growth and other growth conditions such as substrate orientation and growth rate are crucial in persuading high concentrations of silicon to become located on the gallium site and to act as simple substitutional donors. The experimental details of the electrical measurements and the techniques employed to analyse the Fourier data are described in greater detail in reference [1].

3. Carrier concentrations deduced from Fourier analysis of the Shubnikov–de Haas effect

Tables 1 and 2 show the carrier concentrations of the electric subbands deduced from Fourier analysis of the Shubnikov–de Haas data at ambient pressure and as a function of pressure. The total carrier density at zero pressure is estimated to be 7.2×10^{12} cm^{-2} for the 2 nm slab, 8.7×10^{12} cm^{-2} for the 5 nm slab and 1.1×10^{13} cm^{-2} for the 10 nm slab (in the case of the two thinnest slabs the population of the $i = 0$ subband has to be estimated from the theoretical fits, as the Shubnikov–de Haas peaks from this subband were only detectable after a few kilobars of pressure had been applied). These values suggest a loss of 3.8×10^{12} cm^{-2} and 2.3×10^{12} cm^{-2} carriers to localized states for the two thinnest slabs even at ambient pressure if no site switching had taken place. The value found with the thinnest slab agrees quite closely with the saturation value of 5.6×10^{12} cm^{-2} for the carrier concentration achievable with a truly delta-doped sample as estimated in reference [7]. There are two more items of evidence that at least a partial occupation of the localized centres occurs at atmospheric pressure with the two thinnest slabs. The first is that the free-electron concentration decreases immediately on increasing the hydrostatic pressure above atmospheric. The second concerns the persistent photoconductivity effect found after illuminating the sample with a red LED as described in reference [1]. All of these results confirm the LVM conclusion [6] that saturation of the carrier concentration in thin doping slabs for low growth temperatures at a value of 7×10^{12} cm^{-2} is occurring through capture into localized resonant states rather than by site switching of the silicon atoms.

The relative subband occupancies are sensitive to the Fermi level pinning remote to the well and hence to the background doping and to the nature of the defect centres present in the bulk. The sensitivity of the theoretical fit to the choice of depletion charge may provide the major limitation to the accuracy whereby such studies can be employed to study dopant diffusion.

It is worth commenting that 7×10^{12} cm^{-2} distributed over a monolayer would result in an effective volume concentration of 2.6×10^{20} cm^{-3} if no dopant spreading occurred. This is an order of magnitude greater than the limit for incorporation of silicon as a donor even

Table 1. Population of subbands at ambient pressure estimated from Fourier analysis and from self-consistent calculations (all values $\times 10^{12}$ cm^{-2}).

i	2 nm		5 nm			10 nm	
	Expt.	Theor.	Expt.	Theor. (no depletion)	Theor. (depletion)	Expt.	Theor. (depletion)
0	[4.6]	4.62	[5.2]	5.07	5.21	5.35	5.32
1	1.84	1.82	2.32	2.30	2.34	3.28	3.18
2	0.71	0.80	0.90	1.00	0.90	1.66	1.53
3		0.28	0.26	0.40	0.06	0.63	0.67
4		0.08		0.12			0.25
5				0.01			0.06
Total ΣN	7.15	7.60	8.68	8.90	8.51	10.92	11.01
E_{fermi} (meV)		222		219	223		206

[ ] $i = 0$ peak is not observed at ambient pressure with two thinnest slabs, so population is assumed to be the theoretical value.

Table 2. Pressure variation of subband occupancies (all concentrations × 10^{12} cm^{-2}).

Pressure (kbar)	Width (nm)	$i=0$	$i=1$	$i=2$	$i=3$	ΣN_i
	2	[4.6]	1.84	0.71		7.2
0	5 (expt)	[5.2]	2.32	0.90	0.26	8.7
	10	5.35	3.28	1.66	0.63	11.00
6.3	2	4.34	1.38	0.30		6.02
5.1	5(expt)	4.98	2.09	0.72		7.79
5.1	5(th-odp)	4.96	2.22	0.95	0.37	8.50
5.1	5(th-dep)	4.97	2.18	0.77		7.92
6.4	10	5.34	3.46	1.58	0.59	10.92
9	2	4.15	1.30			5.45
10.0	5(expt)	4.25	1.75	0.52		6.52
10.0	5(th-odp)	4.23	1.82	0.74	0.26	7.05
10.0	5(th-dep)	4.23	1.74	0.52		6.49
13.2	2	3.46	0.90			4.36
12.3	5(expt)	3.98	1.58	0.38		5.94
12.3	5(th-odp)	3.94	1.65	0.66	0.22	6.47
12.3	5(th-dep)	3.98	1.59	0.40		5.97
12.4	10	4.56	2.84	1.19	0.36	8.95
15.3	2	3.14	0.73			3.87
16.2	5(expt)	3.14	1.09			4.23
16.2	5(th-odp)	3.05	1.19	0.44	0.12	4.80
16.2	5(th-dep)	3.14	1.13	0.11		4.38
16.3	10	3.76	2.21	0.76		6.73
19	2	1.94				1.94
19.5	10	2.81	1.55	0.42		4.78

th-odp: theoretical value ignoring depletion effects.
th-dep: theoretical value including depletion effects.
expt: experimental values.

at the much reduced growth temperature of 400 °C. Comparisons of published data on Shubnikov–de Haas measurements [8, 9] with those for the sample of 2 nm slab thickness given in this paper and the results of SIMS measurements [10] demonstrate clearly that the silicon is diffusing distances at least of the order of 2–4 nm in supposedly delta-doped samples if the saturation limit is approached with growth temperatures much in excess of 400 °C. In a recent paper Koenraad *et al* [11] report that pressure-induced depopulation is not observed up to 9 kbar with a delta sample grown at 480 °C with a concentration of Si of 8×10^{12} cm^{-2} despite approximately a 50% loss in the free-electron concentration at ambient pressure. The carrier concentration reported for this sample is substantially less than the concentration measured for our thinnest slab at 9 kbar. Thus consistency between the two sets of experiments is achieved if it is assumed that a substantial proportion of the silicon has switched site or condensed into electrically inactive precipitates in the delta sample grown at 480 °C where the volume concentration assuming no spreading is equivalent to 2×10^{20} cm^{-3}. For a carrier concentration of 4×10^{12} cm^{-2} we would not expect to see any depopulation at pressures up to 9 kbar because the localized states will remain above the Fermi energy even for a very abrupt silicon profile.

4. Subband mobilities deduced from Fourier analysis of the Shubnikov–de Haas peaks

The half-width at half-height of each peak in the Fourier spectrum can readily be shown to be given by [1, 12]

$$\delta B_i = \sqrt{3}/2\,\mu_i$$

where μ_i is the mobility of the ith subband.

The results shown in table 3 are a compilation of a detailed analysis of the width and amplitudes of the Shubnikov–de Haas peaks and of a two-carrier analysis of the monotonic magnetoresistance. These show that the mobility in the $i=0$ subband is a factor of three lower than that of the $i=1$ subband and may be an order of magnitude lower compared with the higher-order subbands.

With the thinnest sample at the highest pressure (19 kbar) only a single subband is occupied as shown by a single Shubnikov–de Haas series. Under these conditions the carrier concentrations determined by the different experimental techniques and the Hall mobility should agree to within experimental error as should the Hall and Shubnikov–de Haas results for the carrier concentration. Agreement to within about 10% is indeed achieved (table 4).

Table 3. Subband mobilities deduced from Fourier analysis and galvanomagnetic measurements (all values in cm^2 V^{-1} s^{-1}).

	Pressure (kbar)	μ_0	μ_1	μ_2	μ_3
2 nm sample	0	600	1800	2900	
	6.3	950	2600	4000	
	9	1100	2900		
	13.2	1300	3100		
	15.3	1400	3500		
	19	1400			
5 nm sample	0	600	1100	2600	
	5.1	750	1500	3700	
	10	1000	1900	3900	
	12.3	1200	2300	5000	
	16.2	1500	2900		
10 nm sample	0	650	800	1400	2900
	6.4	900	1000	1700	3900
	12.4	1250	1350	2300	5800
	16.3	1500	1700	3900	

Table 4. Mobility and carrier concentration of the 2 nm sample at 19 kbar from different experimental techniques.

	Shubnikov–de Haas	Hall
Mobility (cm^2 V^{-1} s^{-1})	1400	1448
Carrier concentration (10^{12} cm^{-2})	1.94	1.83

Table 5. Hall and conductivity mobilities (measured and calculated values) (all mobilities in $cm^2 V^{-1} s^{-1}$).

	Pressure (kbar)	μ_{Hall} (meas.)	μ_{Hall} (calc.)	$\mu_{conduct}$ (meas.)	$\mu_{conduct}$ (calc.)
2 nm sample	0	2500	1700	1900	1200
	6.3	2500	2000	1700	1500
	9	2200	1900	1700	1500
	13.2	2200	2000	1800	1700
	15.3	1900	2200	1700	1800
	19	1450	1450		1450
5 nm sample	0				
	5.1	2400	1800	1900	1200
	10	2500	1900	2100	1500
	12.3	2400	2500	2100	1800
	16.2	2200	2000	2100	1900
10 nm sample	0	1900	1300	1700	900
	6.4	2000	1600	1700	1200
	12.4	2100	1700	1900	1600
	16.3	2400	1800	2200	1700

These values are used to estimate the pressure dependence of the Hall and conductivity mobilities and are compared with experiment in table 5. The qualitative agreement is good. For example it can be seen that with the thinnest slab the Hall and conductivity mobilities actually fall at the highest pressure although the mobilities in *all* the individual subbands increase monotonically with increasing pressure. This striking qualitative difference simply arises from the multicarrier nature of transport in the thin doping slabs. The depopulation of the high-mobility higher subbands with increasing pressure causes the relative occupancy of the low-mobility $i = 0$ subband to increase. The Hall mobility, which contains a weighted average of the mobility of all subbands, can therefore fall even if all the subband mobilities are increasing. It should also be noted that Yamada and Makimoto [13] report qualitatively very similar results with a rapidly increasing mobility with increasing subband energy. It should be stressed that the carrier concentrations from Hall measurements *without multiple carrier analysis* may be grossly in error because of the large differences in mobility in the different subbands. Because of the large differences in mobility for the individual subbands, the Shubnikov-de Haas peaks will always be superimposed on a large monotonic magnetoresistance background in delta-doped samples or thin doping slabs except in the case when only one subband is occupied.

The relative mobilities for different subbands in a two-dimensional electron gas (2DEG) have mostly been studied in various heterostructure systems [14–19] with the general conclusion that the mobility in the lowest subband is higher than that in the other bands, although the opposite case has also been reported [14]. In contrast, as mentioned above, work on a delta-plane doping sample has concluded that the mobility increases with increasing subband index [13]. These observations can be understood in terms of an interplay between (i) the physical extent and separation of the electron distribution from the ionized donors, which increases with increasing subband energy, giving reduced scattering, and (ii) the Fermi velocity of the carriers, which is lower in higher subbands, resulting in increased scattering. In single heterojunctions, in which all the electron distributions are remote from any ionized scatterers, it is the second factor which usually dominates, although as Mori and Ando [20] have calculated, it is possible for μ_1 to go from less than μ_0 (factor (ii) dominant) to above μ_0 (factor (i) dominant) as the carrier density in the 2DEG is increased by illumination. (It has been pointed out in [19] that the wavelength of the illumination used can affect the extent of the electron distribution In the delta wells, however, where the ionized donors are to be found in the centre of the well itself ($z = 0$), one notes that the electron probability of the lowest subband is centred in the well plane, while the next excited state has a node at this location. Higher excited states, whether or not they have a node at $z = 0$, have most of their weight even further from the region of dopants centred at $z = 0$.

Also, the electron concentrations in delta wells are generally higher than those in single heterostructures, so that the relative Fermi velocities in adjacent subbands are closer, making this a less important factor. The net result is thus a mobility which increases with subband index.

Since the lowest subband in delta wells is the one in which the electrons are confined to a region close to the ionized donors, a decrease in the donor density should have a strong effect on the ionized impurity scattering which limits the mobility. Electrons in the first excited state will be relatively unaffected by this change both because of their physical separation from the scatterers and because of the shielding of the donors by the electrons in the lowest level.

This change in the relative subband mobilities can be understood qualitatively in terms of a simple picture in which the effect of an increase in pressure is to allow condensation of some of the mobile electrons in the well onto localized states associated with the Si donors whose ionized states form the well itself. One is therefore comparing subband mobilities of two different systems, characterized by a higher (ambient pressure) or a lower (high pressure) net doping density. Furthermore when the dopants are spread into a slab, the donors located at the centre of the slab may be preferentially neutralized.

Another effect also operates to increase the relative mobility of the lowest subband when the donor density of the well is effectively reduced. Higher effective doping (a stronger well) acts to squeeze the wavefunctions of the various subbands into closer proximity to the doping plane. For a many-subband system, the lowest subband spread is relatively independent of the well strength and is limited instead by the effective Bohr radius, which decreases with pressure because of the increase in effective mass and the decrease in dielectric constant. When only one or two excited levels are present, however, a decrease in well strength can have an important effect on the wavefunction of the lowest level. In particular, if we consider a delta well with a uniform doping width of 2.2 nm, and two different doping densities typical of this experiment, namely 10^{13} cm^{-2} (the case for ambient pressure) and 4×10^{12} cm^{-2} (typical of the net doping of the same sample at high pressures), we find that the full width at half maximum of the lowest subband probability distribution is only 4.5 nm in the former case, compared with 6.2 nm in the latter. Thus, at these doping levels in GaAs, a decrease in the effective well strength not only reduces the number of scattering centres seen by the electrons in the lowest subband, but also reduces the probability that these will see the scatterers that remain. These factors are partially offset by the increased scattering probability due to the lower Fermi velocity.

The first calculations of the mobilities in different subbands have recently been reported by Mezrin and Shik [21] who developed a quasi-classical theory in the high-density limit. For 1×10^{13} cm^{-2} Si donors forming a delta spike in GaAs the mobilities in the first three subbands are predicted to be 1350, 3300 and 4800 cm^2 V^{-1} s^{-1} which are substantially larger than the experimental values given in table 4 for the thinnest sample. However, Mezrin and Shik [21] point out that the mobility derived from the Shubnikov–de Haas effect may be different from that measured in the limit of weak fields. Such a difference was found empirically for the Shubnikov–de Haas effect in bulk InSb [22] where the low-field mobility was a factor of 3 greater than that found from the Dingle temperature derived for the highest-field Shubnikov–de Haas peaks. Merzin and Shik also expect the mobility derived from the Shubnikov–de Haas amplitudes for delta-doped samples to be substantially less than the values quoted above, but only expect some 30% difference between the mobility in the ground state and those in the excited states.

Experimentally the pressure measurements show an increase in the mobility for all subbands and only a small relative increase for the lowest subband as the sample is subjected to high pressures. An increase in effective mass of 11.8% and decrease in dielectric constant of 3.3% are expected at 20 kbar from the results reported in reference [23]. From the results presented by Merzin and Shik the mobility in the $i = 0$ subband is expected to be 1120 cm^2 V^{-1} s^{-1} at 20 kbar, i.e. the theory predicts a small fall in the subband mobility on applying pressure in contrast to the greater than factor of two increase observed experimentally. It is too early to speculate whether this difference between theory and experiment arises from correlation effects or for other reasons. Qualitatively the behaviour observed experimentally is very similar to that reported by Maude *et al* [2, 3] for bulk samples. Thus, with a bulk sample doped with 1.2×10^{19} cm^{-3} silicon atoms, the electron concentration drops to 40% of the doping value at a pressure of 16 kbar and the mobility increases by about 1.5. For an Sn sample doped to 1.8×10^{19} cm^{-3} the carrier concentration drops to 40% of the initial figure and the mobility increases by 2.6. The figures for the 10, 5 and 2 nm samples are 60%, 40% and 32% for the carrier concentration and 2.6, 2.9 and 2.3 for the mobility changes respectively. Any changes in mobility are rather similar for different subbands. Furthermore the pressure dependence of the subband occupancies for the 5 nm slab sample can be fitted well by theory which assumes a uniform distribution of positive charge in the slab and a small amount of depletion charge, as can be seen from table 2.

5. Conclusion

We can see little evidence for correlation effects in our analysis of the Shubnikov–de Haas results and hence favour the interpretation that the final charge state of the silicon donors on capturing the electrons is neutral. However, as reported in references [1] and [24] we have evidence that a proportion of the localized states are also non-metastable and this could reduce the proportion of D^+D^- pairs formed. In addition the deliberate spreading of the donors over a finite slab to avoid site switching or the formation of precipitates rather than the deposition of the silicon onto a single plane may have acted to reduce the magnitude of correlation effects. It is intended to refine the analysis and to extend the measurements to thinner slabs in order to improve the detection limit for correlation effects.

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