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Study of impurity states in a *n–n* GaAs–AlGaAs heterojunction using cyclotron resonance and Shubnikov–De Haas experiments[1])

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Abstract. The electronic structure of two *n–n* GaAs–AlGaAs heterojunctions is studied using far-infrared cyclotron resonance and Shubnikov–de Haas effect. Shallow impurity states, similar to the so called "hydrogenic" impurities, are found in both systems. It is found that these impurity states lead to a freezing out of electrons from the conduction band with decreasing temperature.

I. Introduction

Two dimensional (2D) electron systems have attracted a lot of interest in recent years. Most of the investigations on these systems have centred on heterojunctions (MOS, SS) and superlattices [1]. These investigations have given considerable insight into the fundamental properties of 2D systems, and have been of great importance in the development of new electronic devices.

The most useful technique for the study of these systems is cyclotron resonance (CR) and the Shubnikov–de Haas effect (SdH). Among the great number of papers published in this field, studies of CR linewidth, mass analysis as well as quantum oscillations in tilted field [2] have proven to be a powerful tool in understanding the 2D electron-interface impurity system.

These same techniques have also been used to study questions on the related problem of activated conduction in Si inversion layers [3]. However it should be noted that one of the difficulties in characterizing the Si–SiO_2 interface is caused by the presence of rather ill defined surface-imperfections. A more promising system to study is the GaAs–AlGaAs heterojunction. Here, because of the excellent lattice match, the interface impurities which are believed to be present

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[1]) In honour of J. L. Olsen's 60th birthday.
[2]) Dipl.Phys. ETH, 1979 (with J. L. Olsen as supervisor).
[3]) Dipl.Phys. ETH, 1959 (with J. L. Olsen as supervisor).

are mainly bulk like. To a large extent investigations on GaAs–AlGaAs have centred on n–p heterojunctions. In this system, the interaction between impurities and 2D interface electron states are small. In comparison, the impurity-interface interaction in n–n heterojunctions is large and may lead to activated conduction.

In this paper we describe CR and SdH experiments on two n-type GaAs–AlGaAs heterojunctions where Hall measurements suggest an activated conductivity at temperatures below 77 K. The far-infrared (FIR) data are found to differ markedly from those of Si inversion layers [4] and n–p GaAs–AlGaAs heterojunctions. Here, no localization in the band edge seems to occur. We must conclude that the cause of the activated conductivity is due to shallow donor impurities present in the GaAs layer. These shallow donor impurity states are very similar to the so called "hydrogenic" impurities found in the bulk for a variety of III–V compounds [5]. We believe that this is the first observation of a freezing out of 2D electrons in the accumulation layer of a heterojunction due to shallow n-type impurity states.

II. Sample preparation and experimental set-up

The samples were grown using liquid phase epitaxy into a (multi) layer structure of alternating GaAs and $Al_xGa_{1-x}As$ ($x = 0.3$) layers. The GaAs is unintentionally doped with a final donor concentration of $\sim 1.10^{15}$ cm^{-3}. Sample 430 has 3 GaAs and 4 AlGaAs alternating layers with a nominal thickness of 1430 Å per layer, while sample 456 has only a single layer of GaAs and AlGaAs with a nominal thickness of 10000 Å and 1500 Å respectively. The substrate is semi-insulating GaAs with Cr as a dopant. The samples were of a suitable geometry for van der Pauw and SdH measurements using a 4-point low frequency a.c. technique.

The cyclotron resonnace (CR) measurements were performed in the Faraday configuration using far-infrared radiation obtained from an optically pumped (CO_2) CH_3OH laser [6]. The measurements were performed in a 15 tesla Bitter magnet at the High Field Magnet Laboratory of the University of Nijmegen [7]. The transmission spectra were measured using FIR produced by a Grubb–Parsons Michelson interferometer [8].

III. Experimental results and discussion

(a) *Characterization of the samples using cyclotron resonance and* d.c. *transport measurements*

The far-infrared transmission of samples 430 and 456 is shown in Fig. 1 for incident radiation of $\lambda = 57$, 70 and 118 μm at temperatures of 6, 35 and 77 K as a function of magnetic field. The lower field transmission minima have been identified as a $1s$–$2p$, [5] and the high field transmission minima can be identified with CR.

The peak position of the resonance fields remain constant as a function of temperature, they show a $B_{CR} = (\omega m^*/e)$ and $B_{1s-2p} = B_{CR} - (m^*/\hbar e)\Delta E$ dependence (with $m^* = 0.071 m_0$ and $0.070 m_0$, with $\Delta E = 3.9$ meV and 3.6 meV for the

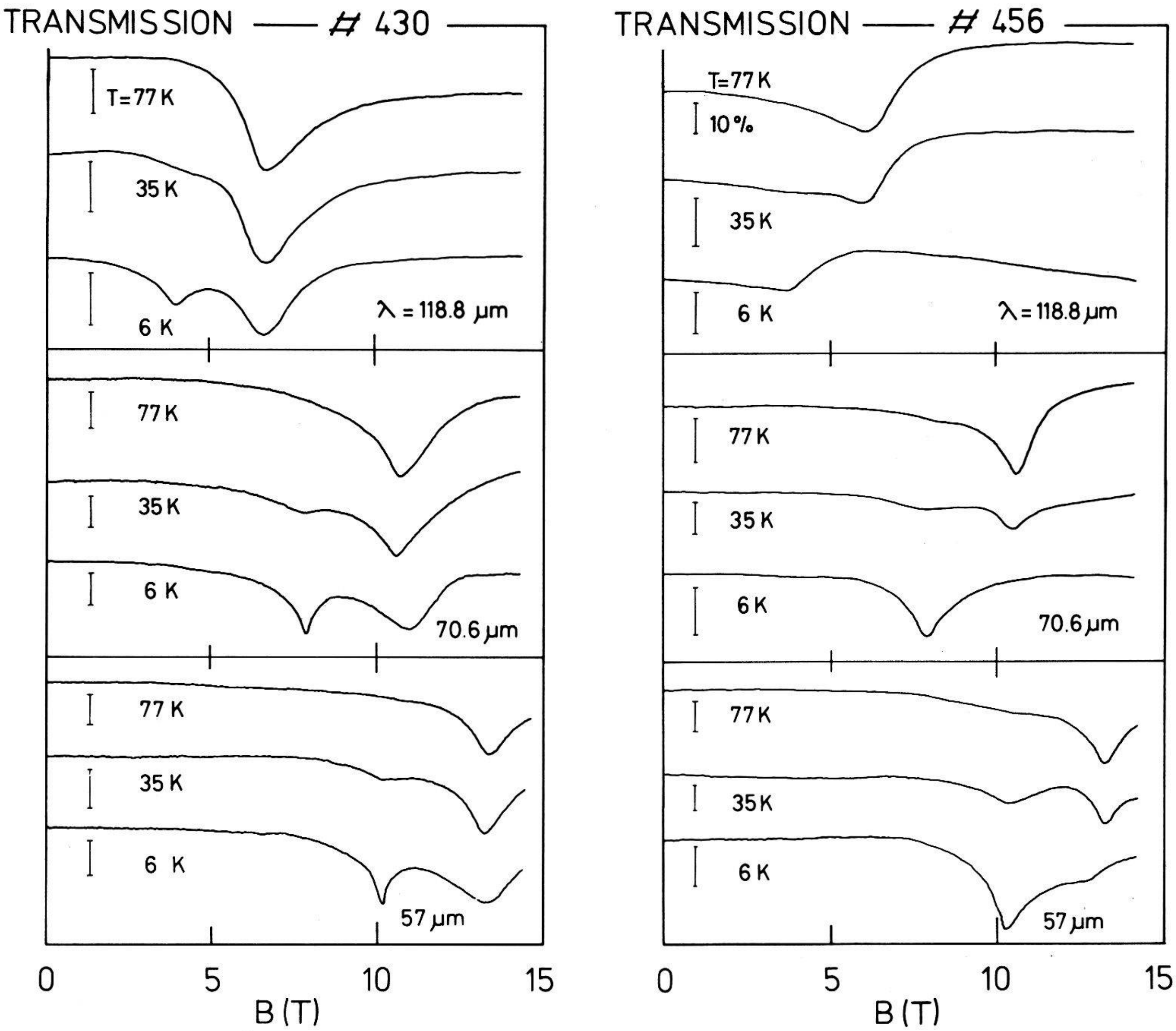

Figure 1
The FIR-transmission of the multilayer n-GaAs–AlGaAs heterojunction sample 430 and the single layer n-GaAs–AlGaAs heterojunction sample 456 for various temperatures and different wavelengths of the incident radiation as a function of the magnetic field. The vertical bars indicate a change in transmission of 10%.

samples 430 and 456 respectively). This is illustrated in Fig. 2. What seems generally true for the two samples is that on lowering the temperature, the amplitude of the $1s$–$2p$ transition increases as the CR amplitude decreases. This is more pronounced for sample 456: for example in Fig. 1 (sample 456), the $1s$–$2p$ transition seems to grow at the expense of the CR transition to the extent that at the lowest temperature measured ($T = 6$ K) there remains no CR minima for $\lambda = 118$ and 70 μm. On the basis of CR, SdH and Hall data, we will argue that this decrease of the CR amplitude on decreasing the temperature is due to the fact that interface electrons freeze out at low temperatures in the same way as is known from bulk semiconductors. We have indications that this freeze-out occurs not in all of the 6 interface layers of the multilayer sample 430 which explains the quantitatively different behaviour of the two samples investigated.

In order to make a more quantitative study of the CR data, we define the

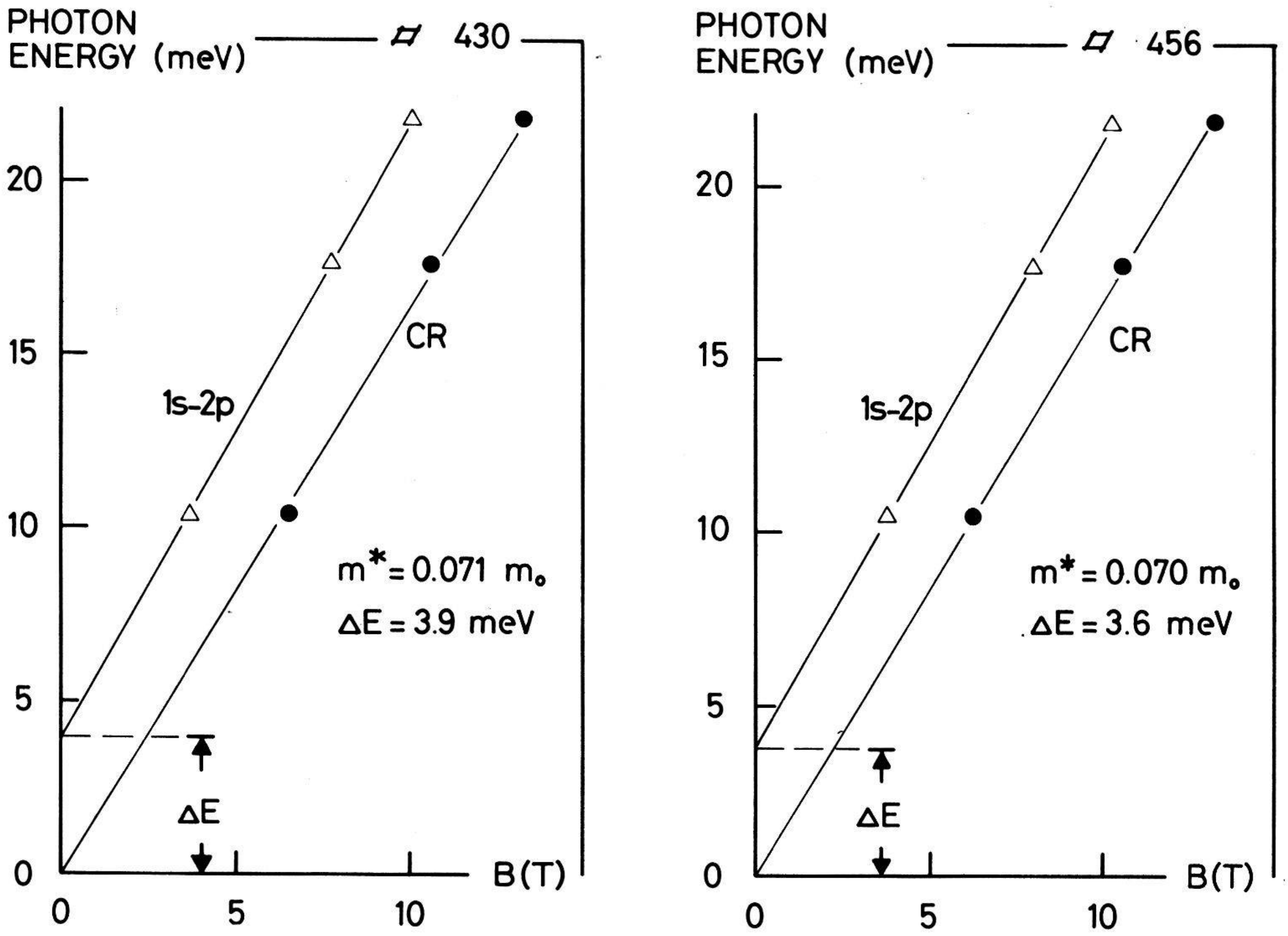

Figure 2
Energy of incident radiation as a function of peak field for the heterojunctions. 1*s*–2*p*: impurity level signal. CR: cyclotron resonance signal.

absorption power AP of a transmission minimum to be the strength of the minimum times the half width at full maximum. The average value of AP at 70 μm and 118 μm is determined in this way and shown as a function of temperature in Fig. 3. The AP of a CR transmission signal is expected to be proportional to the number of conduction electrons times a slowly varying function of the temperature. This can be seen in Fig. 3. Note however, that the AP of sample 456 tends to zero for low temperatures while that of sample 430 remains finite. At 77 K, one gets for the ratio $AP_{430}/AP_{456} \simeq 4$ instead of 6 as one would expect from the number of interfaces in both samples, assuming that all the electrons are confined to the interfaces; this indicates that at 77 K bulk electrons also contribute to the AP. Assuming a constant AP_{2D} across one interface and a uniform AP_{3D} per unit length in the layer, from the numbers of interfaces and thicknesses of the GaAs layers one can estimate that at 77 K the bulk contribution is about 1/3 for sample 456 while it is about 1/130 for sample 430. On lowering the temperature the bulk contribution will disappear and the difference in the AP for the two samples is determined by the number of the remaining electrons at the interfaces only.

This freeze-out of the 2D electrons is also reflected in the d.c.-transport measurements (Table 1). The electron sheet density per interface $n^s_{\mathrm{Hall}} = (n_e d)/s$

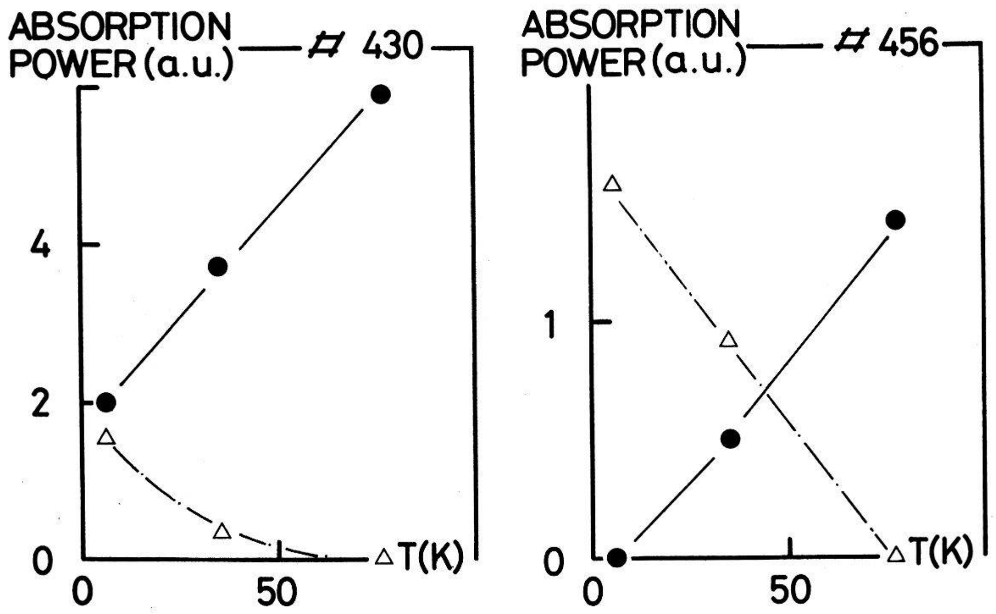

Figure 3
Absorption power as a function of temperature. The triangles ▲ denote $1s$–$2p$ transitions and the full circles ● denote the CR transitions. The lines are drawn to guide the eye.

(with s the number of GaAs–AlGaAs interfaces) and mobility μ_{Hall} are deduced by Hall measurements at temperatures of 300, 77 and 4.2 K. The sheet density n^s_{SdH} and effective mass m^* are obtained by measuring the oscillation period and temperature dependence of the SdH signal. The Fermi energy E_F is derived from $E_F = n^s_{\text{SdH}}/D(E)$ with $D(E) = 2m^*/\pi\hbar^2$, the density of states of a 2D electron gas, assuming a spin degeneracy of 2. For the multiheterojunction sample 430, with 6 interfaces, one finds different values for the electron sheet densities deduced from Hall or SdH measurements, i.e. at 4.2 K n^s_{Hall} is about 4 times larger than n^s_{SdH}.

Table 1
Sample parameters deduced from transport measurements

	Sample 430 (3GaAs, 4AlGaAs alternating layers, $d = 1430$ Å per layer)		Sample 456 (GaAs–AlGaAs single layer, $d_{\text{GaAs}} = 10000$ Å, $d_{\text{AlGaAs}} = 1500$ Å)	
T(K)	n^s_{Hall} (cm^{-2})	μ_{Hall} (cm^2/V-sec)	n^s_{Hall} (cm^{-2})	μ_{Hall} (cm^2/V-sec)
300	2.2×10^{12}	4940	1.6×10^{12}	5360
77	1.2×10^{12}	14820	1.1×10^{12}	13900
4.2	1.2×10^{11}	16330	—	~430†
	n^s_{SdH} (cm^{-2})	E_F (meV)	n^s_{SdH} (cm^{-2})	E_F (meV)
4.2	4.7×10^{11}	16	4.4×10^{10}	1.5‡
	$m^* = 0.071\ m_0$		$m^* = 0.070\ m_0$‡	

†) Deduced from SdH.
‡) Deduced from CR.

This can be interpreted as an indication that not all the interfaces behave in the same way, in some the 2D electron system may be frozen-out and in others not. The SdH data of this sample show one single oscillation, periodic in $1/B$, from which the sheet density of each of the non-frozen-out 2D layers is determined to be $n^s = 4.7 \cdot 10^{11}\ \mathrm{cm}^{-2}$. The SdH effective mass $m^* = 0.071 m_0$ is in excellent agreement with the one found from CR. It may then be assumed that the remaining interfaces of sample 430 behave like the interface of the single layer sample 456. There, at 4.2 K only a weak SdH signal is measured with an oscillation period corresponding to $n_s = 4.4 \cdot 10^{10}\ \mathrm{cm}^{-2}$. Unfortunately, the weak SdH signal in combination with the high resistance at 4.2 K prevents an extraction of a reliable value for the effective mass m^* or the Hall electron sheet density. However, using the effective mass $m^*_{CR} = 0.070 m_0$ determined by CR measurements, it is now possible to determine the Fermi energy for this sample to be $E_F = 1.5$ meV. From the sheet conductivity at zero field and the SdH sheet density n^s_{SdH} one can estimate a value for the electron mobility $\mu \simeq 430\ \mathrm{cm}^2/\mathrm{V} \cdot \mathrm{sec}$. The simple fact that CR has been observed at 6 K for $\lambda = 57\ \mu$m with a line width comparable to that at 35 and 77 K indicates that this low-field value for the mobility is not representative of the mobility in high magnetic fields. It is possible that the strong localization present at low temperatures is partially removed by the high magnetic fields. This fact may also be the origin of the increased AP of the CR signal for $\lambda = 57\ \mu$m with respect to that of $\lambda = 70\ \mu$m and 118 μm observed at low temperatures in both samples. From the SdH data of the multilayer sample 430 no direct evidence for the existence of frozen-out interface electron layers is found; this may be due to the fact that the strong SdH signal due to the non-frozen-out layers overshadows a possible weak signal (as seen in sample 456). However, from the shape of the CR signal at 6 K for $\lambda = 57\ \mu$m two different absorption peaks can be resolved and be attributed to both types of interfaces. The shape of the CR absorption peak at 6 K for $\lambda = 57\ \mu$m for sample 430 is asymmetrically broadened at the low field side of the peak field. In contrast, the CR shape for the single layer sample 456 at the same temperature and wavelength is symmetric, but shows a remarkable decrease of the peak field of about 2.5% with respect to the high temperature value. Therefore, the CR absorption peak of sample 456 at 6 K can be understood to be due to two different kinds of 2D electron layers; the first is a degenerate electron gas with $E_F = 16$ meV, and the second a nearly frozen-out electron gas with a very low Fermi energy. The observed decrease in peak field $B_{CR} = (\omega/e) m^*_{CR}$ must be attributed to a decrease of m^*_{CR} of 2.5%. This can be explained due to band-nonparabolicity accompanied by a decrease of the Fermi energy of about 16 meV. It is interesting to note that at temperatures above 77 K the Hall data of both samples are very similar. No freeze-out appears and all the interfaces seem to carry electrons. Finally, the number of "good" 2D interface layers (i.e. layers carrying electrons at low temperatures as well) in the multilayer sample 430 can be estimated from transport and CR measurements independently. Defining s' the number of "good" 2D layers, we get from the SdH and the Hall sheet density $s' = 6 n^s_{Hall}/n^s_{Hall} \cong 1.5$. From the CR absorption power we get $s' = \mathrm{AP}\ (77\ \mathrm{K})/\mathrm{AP}\ (4.2\ \mathrm{K}) \cong 2$, assuming a temperature independent absorption. Therefore it seem reasonable to assume that the multilayer sample 430 has two interfaces carrying a degenerate 2D electron gas, and four interfaces showing a freeze-out.

(b) *Far-infrared transmission and donor impurity states*

In view of the clear indications for depletion found in these 2D interface layers under investigation, it is of considerable interest to investigate the impurity levels using spectroscopic means as well. This has been done by measuring the far-infrared transmission in zero magnetic field, by measuring the temperature dependence of the AP of the $1s$–$2p$ transition of the impurity states, and by studying the angular dependence of the CR and the $1s$–$2p$ transition.

Figure 4a shows the angular dependence of the CR and the $1s$–$2p$ transition for sample 430. The CR signal shows the typical 2D behaviour, i.e. the CR peak field follows closely a $B_0/\cos\theta$-curve (full line in Fig. 4b), with θ the angle between the direction of the magnetic field and the normal to the layers. This same $B_0/\cos\theta$ dependence is also seen in the SdH measurements of both samples. In contrast, the angular dependence of the $1s$–$2p$ signal indicates, that similar to the spin Zeeman effect [9] the total magnetic field is responsible for the splitting rather than the perpendicular component $B_\perp$ which determines the Landau level splitting.

As the angular dependence allows a differentiation between the 2D and 3D electrons, one can make a crude estimate of an upper limit of a possible

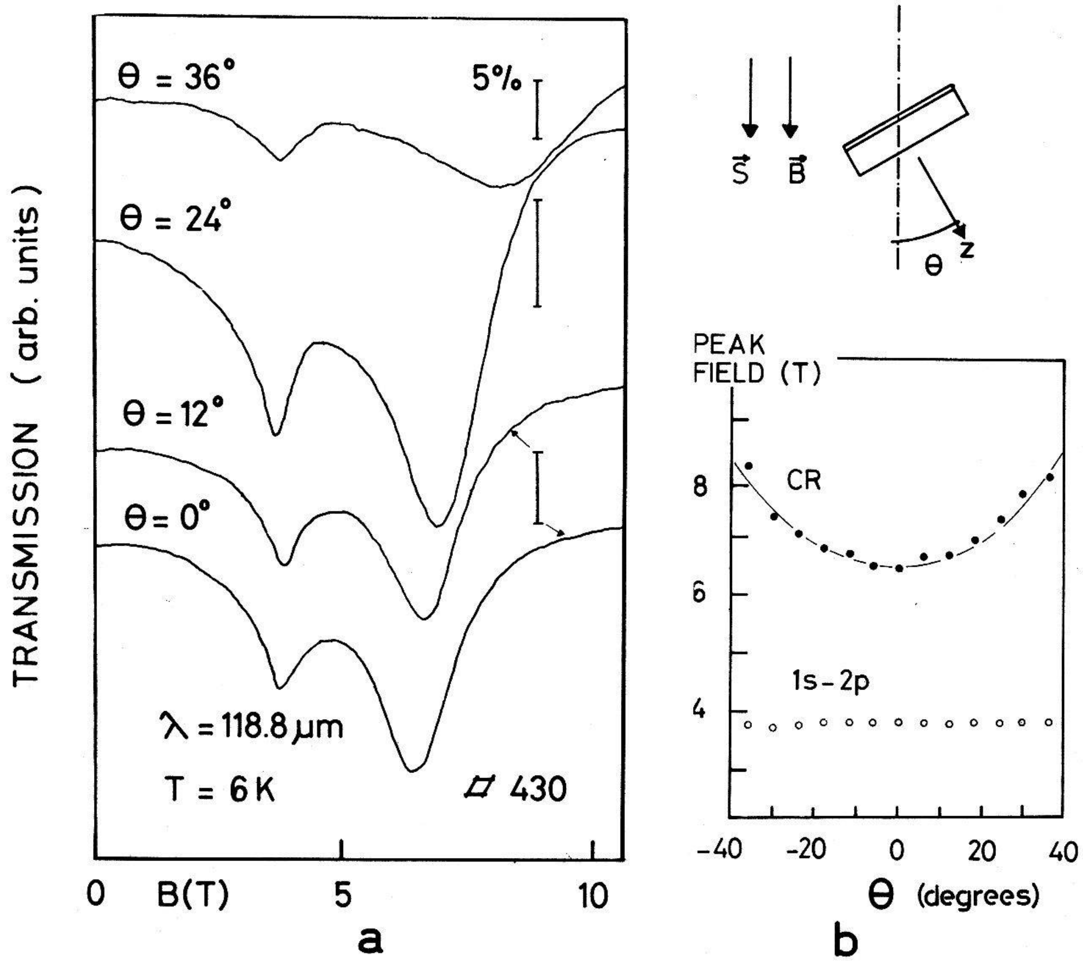

Figure 4
(a) The FIR transmission of sample 430 for various angles. A transmission of 5% is indicated by the vertical bars. (b) The angular dependence of the peak field (CR and $1s$–$2p$) for sample 430. The solid lines represent a $B/\cos\theta$ dependence.

contribution due to 3D electrons: as now 3D signals could potentially be seen at the unrotated position, one gets from the signal to noise ratio in sample 430 at 6 K an upper limit of the 3D sheet density of $n^s_{3D} < 0.5 \cdot 10^{10}\,\text{cm}^{-2}$.

Note that the observed independence of the angle of the $1s$–$2p$ transition does not necessarily mean that the impurities are distributed uniformly through the whole bulk. We will now argue that the impurities are confined at the GaAs side of the interface and that they are probably the origin of the observed freeze-out.

The $1s$–$2p$ transition is considered to be due to shallow hydrogenic donor states. It is well known, that in the high field limit the Zeeman effect of this system is proportional to the effective mass of the conduction electrons of the relevant material. As can be seen in Fig. 3, the observed Zeeman resonance field B_{1s-2p} is proportional to the cyclotron resonance field B_{CR}, which, through the effective mass m^*_{CR}, is in itself associated with the GaAs and not the AlGaAs layer. In addition, it is possible to make a rough estimate of the binding energies of these impurity states. If the field dependence of the energy splitting of the $1s$–$2p$ resonance is linearly extrapolated to zero field (see Fig. 2), one gets a value of $\Delta E = 3.9$ meV for sample 430 and $\Delta E = 3.6$ meV for sample 456. using the same linear extrapolation scheme for the bulk GaAs system, one gets $\Delta E_{bulk} = 3.59$ meV. [5] The binding energy is proportional to the zero field splitting

$$\left(E_N = \frac{e^4 m^*}{2(4\pi\varepsilon\varepsilon_0\hbar^2)}\frac{1}{N^2}\right).$$

Using a bulk value for the binding energy in GaAs of 5.86 meV [5], one gets values of 6.3 meV and 5.9 meV for sample 430 and sample 456 respectively. These differences can be attributed to the specific impurity itself or possibly to the interface dipole field.

It is interesting to note that the total absorption power of sample 456 ($AP_{1s-2p} + AP_{CR}$) remains approximately constant, independent of the temperature. In order to analyse this feature, we have performed separate CR measurements on a non-degenerate bulk n-GaAs sample under the same experimental conditions. These measurements show that the transition probabilities for the $1s$–$2p$ transition and the CR transition are equal, and that the absorption power is proportional to the number of neutral donors (n_D) and conduction electrons (n_e), determined independently from transport measurements. In analogy to these bulk experiments, we may therefore assume that in these heterojunctions the donor states are filled up at the expenses of the 2D conduction electrons, and that the total number of donors is the same as the number of electrons at 77 K ($n_e d = n_D d = 1.1 \cdot 10^{12}\,\text{cm}^{-2}$ for sample 456). Using this number and assuming a uniform distribution of the impurities over the 10000 Å thick GaAs layer, one gets an impurity concentration of $n_D \simeq 1.1 \cdot 10^{16}\,\text{cm}^{-3}$. This value is about a factor of 10 higher than expected from the donor concentration deduced using bulk test specimens grown under the same conditions. Therefore, we expect that a considerable amount of donors is introduced into the GaAs of the heterojunction by cross diffusion from the highly doped AlGaAs layer and not during the GaAs growth cycle.

In an attempt to characterize the donor states of these heterojunctions more directly, we have also measured the far-infrared transmission spectrum for sample

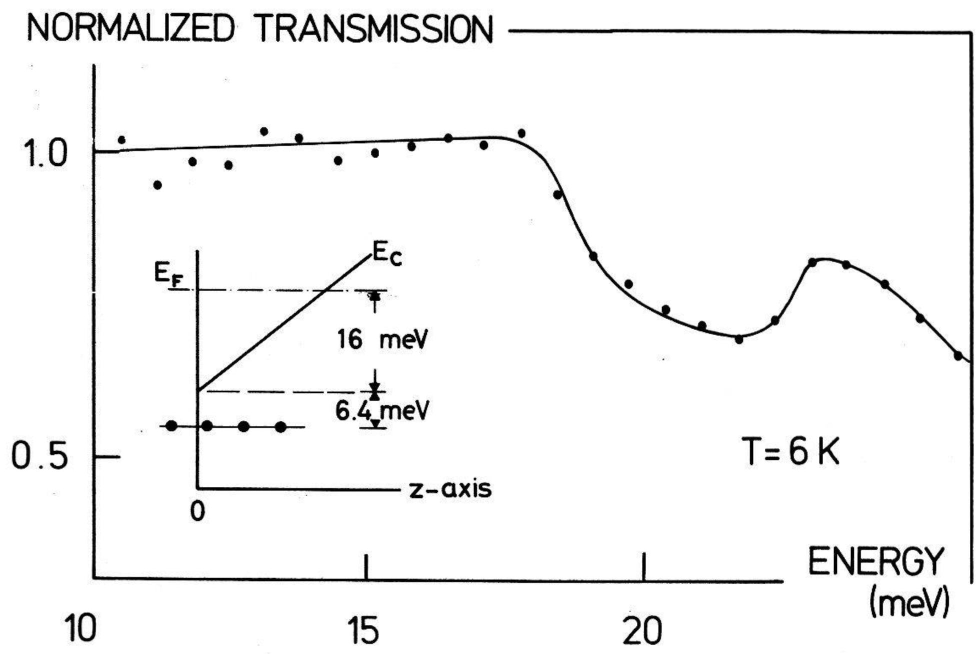

Figure 5
Normalized transmission of sample 430 with respect to bulk GaAs as a function of incident radiation; the insert indicates the schematic energy band at the interface ($z = 0$).

430 as well as for a bulk n-GaAs reference sample. The transmission of sample 430, normalized with respect to bulk n-GaAs is shown in Fig. 5. At low energies, the normalized transmission is nearly constant and arbitrarily adjusted to unity; however, near 20 meV a minimum appears which is not present in bulk GaAs and which is therefore related to the heterojunction. In order to investigate whether this minimum is due to possible interface effects inherent to such heterostructures (e.g. interface states), we also measured the transmission of a similar n–p-multiheterojunction, where no freeze-out has been found; here, the spectrum is found to be identical to the one of bulk GaAs in the energy region investigated. As the difference of this multilayer compared with sample 430 is the growth temperature (leaving the GaAs-layer slightly p-doped), we may conclude that the absorption minimum of the n-type sample 430 is due to a shallow donor absorption and not due to interface states. This broad absorption at about 20 meV can be identified with a transition from the 1s-shallow impurity level to states in the conduction band above E_F; this is in fair agreement with the value of $E_F = 16$ meV and an impurity binding energy of 6.3 meV as found in sample 430. Note that this transition can only occur at the interface (and not in the bulk) as is shown schematically in the insert of Fig. 5.

IV. Conclusions

In a non-degenerate n-type bulk semiconductor a freezing-out of the electrons to the donor states is well known from the literature; in the single layer and multilayer heterojunction investigated in the present study, we now also clearly see a freeze-out of the 2D conduction electrons to impurity states as the temperature decreases. The angular dependence of the CR-signal, SdH measurements, the existence of high mobility coupled with a high sheet density $n_e \cdot d$

(d = thickness of the GaAs layer) and the mass enhancement shows that the electrons are 2D confined. If present, the number of bulk conduction electrons must be quite small ($n_e \cdot d < 0.5\ 10^{-10}\ \mathrm{cm}^{-2}$). A strong absorption peak at 20 meV is identified with a transition from an impurity level to the conduction band at the interface. The mass enhancement of 4% relative to bulk GaAs must be due to either small band bending or a wide potential well and suggests a smooth AlGaAs–GaAs interface. An enhanced CR-absorption in high magnetic field could indicate a delocalization of confined electrons, but unfortunately no further evidence in transport measurements has yet been found.

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