Cyclotron-resonance study of nonparabolicity and screening in GaAs-$Ga_{1-x}Al_xAs$ heterojunctions

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Cyclotron resonance has been used to study the frequency, electron-concentration, and temperature dependences of the effective mass in GaAs-$Ga_{1-x}Al_xAs$ heterojunctions. In samples of low carrier concentration the energy dependence of the low-temperature mass has been measured in the quantum limit, and the variation of the mass with Fermi energy at low fields has been studied in samples of various electron concentrations and by the use of persistent photoconductivity. The results give an energy dependence of the effective mass which is ~20% lower than that measured in bulk GaAs and which is accounted for by five-band **k**·**p** theory only, consistent with a dramatic reduction in the polaron coupling due to screening. Further evidence of the importance of screening comes from the temperature dependence of the effective mass, which shows a rapid increase of order 2% as the temperature is raised to ~100 K. This is attributed to the reappearance of the polaron coupling as the screening is reduced by thermal broadening of the carrier distribution.

I. INTRODUCTION

The conduction-band structure in a polar semiconductor is modified at low energies by two effects: interband coupling (nonparabolicity) and electron-phonon interactions (polaron coupling). In weakly polar III-V compound semiconductors these effects are of comparable size and there has been considerable interest in measuring and calculating them in both bulk materials and two-dimensional (2D) systems. Bulk GaAs has been extensively studied by cyclotron resonance (CR)[1–3] and it is generally accepted that there is a significant effective-mass (m^*) enhancement from polaron effects, even at energies well below the resonant regime ($\omega_C \simeq \omega_{LO}$). However, the results in 2D systems are less conclusive, since recent studies have shown both increased[4,5] and reduced[6–8] polaron mass enhancements relative to the bulk case. Calculations suggest[9–11] that the reduced dimensionality should result in an almost threefold increase of the polaron coupling over bulk (3D) systems, but there is little evidence for this, and it has been suggested that this enhancement may be quenched by screening in the highly degenerate 2D electron gas.[8,12–14] The finite extent of the wave function in the third dimension has also been shown to reduce this coupling.[8,10,12,15] In this paper we present the results of cyclotron resonance measurements on several very high quality GaAs-$Ga_{1-x}Al_xAs$ heterojunctions in which these effects are studied in detail. The extremely narrow linewidths have allowed accurate measurements to be made of the energy, temperature, and carrier-concentration dependences of the effective mass. The results show that the total nonparabolicity in this system is reduced relative to the bulk material at low temperatures, while the mass shows an anomalous temperature dependence. These effects are attributed to changes in the polaron coupling.

II. EXPERIMENTAL PROCEDURE

The experiments were carried out on several GaAs-$Ga_{1-x}Al_xAs$ heterojunctions grown at Philips Research Laboratories, Redhill, by molecular beam epitaxy.[16] The substrates were semi-insulating GaAs on which high purity p-type GaAs buffer layers were grown, followed by heterojunctions with undoped $Ga_{0.68}Al_{0.32}As$ spacer layer thicknesses between 0 and 800 Å. Cyclotron resonance was detected by measuring the transmission by the sample of radiation from an optically pumped far-infrared laser as a function of magnetic field (B), and the effective mass was deduced from the resonant field. A typical experimental recording is shown in Fig. 1, indicating the extremely narrow linewidths obtained. The

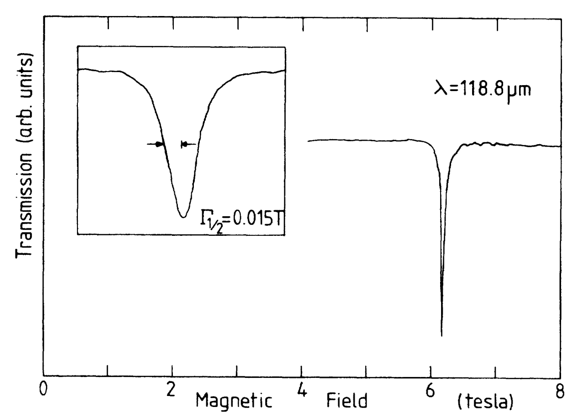

FIG. 1. A typical recording of the transmission of sample G63 at 4.2 K. The linewidth $\Gamma_{1/2}$ is $0.015T$ ($\Delta B/B \simeq \frac{1}{400}$).

effective mass was measured as a function of radiation energy from 2.2 to 29.4 meV, of electron concentration (n) over the range $(0.9-9.1)\times 10^{11}$ cm^{-2}, and of temperature between 1.5 and 280 K. The carrier concentrations were deduced from simultaneous measurements of the Shubnikov–de Haas oscillations in the static conductivity. For a given sample it was possible to vary the electron concentration by a factor of approximately 2 using the persistent photoconductivity brought about by illumination from a red light-emitting diode (LED). For the high-temperature measurements it was found necessary to measure the transmission of a double thickness of the sample and use a circular polarizer to obtain sufficient absorption. The sample substrates were wedged to avoid interference effects.

III. ENERGY DEPENDENCE OF m^*

In a two-dimensional electron gas (2D EG) the degeneracy of each Landau level is $2eB/h$, and the number of occupied levels N is $nh/2eB$, where n is the electron concentration and spin splitting has been neglected. A noninteger value of N indicates that the highest filled level is only partially filled. Above the fundamental field B_F ($=nh/2e$) where $N=1$, only the lowest Landau level is occupied, and at low temperatures the only optical transition which can occur is from the 0 to 1 Landau levels. The average energy of this transition is thus that of the exciting radiation $\hbar\omega_c$ and so the energy dependence of the effective mass may be measured in this region, known as the quantum limit.

At fields below B_F, a number of different transitions may be possible. In the low-field limit the average energy of these transitions remains essentially constant, only oscillating slightly about a fixed energy of $E_F=\hbar^2\pi n/m^*$, and is independent of the transition energy. Thus we would expect m^* to remain almost constant at fields below B_F and to measure the energy dependence of m^* it is necessary to make measurements in the quantum limit.

Three samples with particularly low carrier concentrations (sample G63, $n=0.9\times 10^{11}$ cm^{-2}; sample G29, $n=1.4\times 10^{11}$ cm^{-2}; sample G66, $n=1.4\times 10^{11}$ cm^{-2}) were studied in order to enable the quantum limit to be reached over as large an energy range as possible. Typical sample mobilities for these carrier concentrations were of order 500 000 cm^2/V s. The results from samples G29 and G66 are shown in Fig. 2. The increase in effective mass with energy is found to be approximately linear up to an energy of around 20 meV, above which resonant polaron coupling causes a more rapid increase. This behavior was the same in both samples, which have similar electron concentrations. The results from sample G63 were complicated by strong subband–Landau-level couplings,[17] but were consistent with those from the other two samples. The rate of increase of the mass with energy is substantially less than has been reported recently for the case of bulk GaAs.[3] This result will be discussed later.

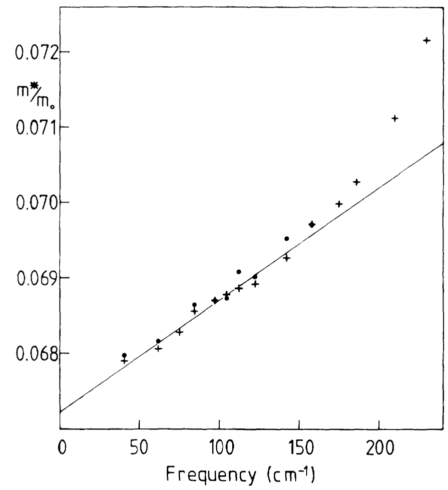

FIG. 2. The frequency dependence of the cyclotron mass at 4.2 K in samples G29 (+) and G66 (●) where $n=1.4\times 10^{11}$ cm^{-2} in both cases. All measurements were made in the quantum limit, where only the lowest cyclotron transition ($L=0$ to 1) occurs. The solid line is a fit to the low-frequency results with a nonparabolicity of $K_2=-1.4$. Above ~ 160 cm^{-1} the mass increases more rapidly as the LO-phonon frequency (296 cm^{-1}) is approached.

All three samples were also illuminated with a red LED to increase their electron concentrations and CR measurements were made in both the low-field and quantum limits. The results from sample G29 before and after strong illumination ($n=1.4\times 10^{11}$ and 3.4×10^{11} cm^{-2}, respectively) are shown in Fig. 3. After illumination the low-field limit corresponds to frequencies below ~ 100 cm^{-1}, and the effective mass remains essentially constant in this region. This was observed in all of the samples studied, in contrast to the results reported by Horst *et al.*,[7] who observed an increasing mass even below B_F.

As discussed earlier, both nonparabolicity and polaron coupling modify the band structure of polar semiconductors. Since these effects are both relatively weak in GaAs it is possible, to a first approximation, to treat the two effects separately and simply add them together. The validity of this assumption is discussed for bulk materials by Das Sarma and Mason,[18] who conclude that it introduces only a few percent error. The band nonparabolicity has been calculated by numerous authors using the **k**·**p** method of Kane.[19] Experimentally, m^* is found to increase linearly with energy at low energies (<20 meV), and so the cyclotron energy can be approximately described by including terms only up to k^4. In such a model considering interactions between three bands, Palik *et al.*[20] derived the relation

$$E(k)=\frac{\hbar^2k^2}{2m^*}+\frac{K_2}{E_g}\left[\frac{\hbar^2k^2}{2m^*}\right]^2, \qquad (1)$$

where K_2 is coefficient determined by the band structure. For a heterojunction this results in an effective

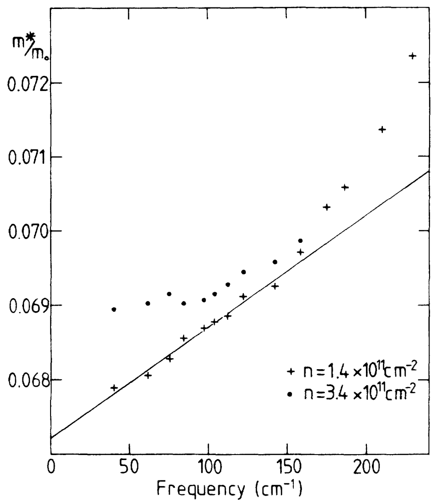

FIG. 3. The frequency dependence of the cyclotron mass in sample G29 before (+) and after (●) increasing the electron concentration by illumination. The mass remains approximately constant until the quantum limit is reached, after which it increases linearly with frequency. At the highest frequencies m^* increases more rapidly as the resonant polaron coupling region is approached. The solid line is a fit to the results with a nonparabolicity of $K_2 = -1.4$.

mass given by

$$\frac{1}{m^*} = \frac{1}{m_0^*}\left\{1+\frac{2K_2}{E_g}[(L+1)\hbar\omega_c + \langle T\rangle_z]\right\}, \quad (2)$$

where m_0^* is the band-edge effective mass, E_g is the band gap, L is the quantum number of the initial Landau level, and $\langle T\rangle_z$ is the kinetic energy due to motion perpendicular to the layers. $\langle T\rangle_z$ is independent of the perpendicular quantizing magnetic field[21] and remains constant if the electron concentration is unchanged. The original three-band calculations[20] give a value of $K_2 = -0.83$ for GaAs. However, recent calculations which include five-band contributions[3,22,23] conclude that the band nonparabolicity is rather larger, and values of K_2 between -1.1 (Ref. 22) and -1.5 (Ref. 23) were derived for GaAs in the low-energy limit. Note that a five-band model is necessary to describe simultaneously the band-edge effective mass, the conduction-band anisotropy away from $k = 0$, and the splitting of the cyclotron resonance due to the two different spin states, as seen in bulk material.[2,3] Equation (2) assumes the band structure to be isotropic.

Polaron coupling results from the interaction of electrons with the polarization field of the longitudinal optic (LO) phonons and is characterized by the Frölich coupling constant α (Ref. 24) which is equal to 0.07 in bulk GaAs. The coupling causes a constant, zero-frequency increase in m^* and an increase in the nonparabolicity, which at low energies ($\omega_C/\omega_{LO} < 0.5$) results in an almost linear increase in m^* with energy until the resonant region is reached. It is consequently very difficult to separate polaron effects from those of the band nonparabolicity (BNP), and the two may be approximately accounted for in Eq. (2) by replacing K_2 with $K_2(\text{tot}) = K_2(\text{BNP}) + K_2(\text{pol})$. Various methods have been used to solve the Frölich Hamiltonian for the 3D case: Rayleigh-Schrödinger perturbation theory[24] (RSPT), valid only for $\alpha < 1$ and $\omega_C/\omega_{LO} \ll 1$, Wigner-Brillouin perturbation theory (WBPT),[25] an improved WBPT, valid only for $\alpha < 0.1$ (Refs. 1, 11, and 26), and variational solutions.[25] However, all of these predict comparable and relatively small values for the nonresonant polaron enhancement of the mass in bulk GaAs, with the largest result being that of Peeters *et al.*,[26] who have included interactions with all higher Landau levels to give results which may be approximated to $K_2(\text{pol}) = -0.4$ in three dimensions and $K_2(\text{pol}) = -1.0$ in the 2D case for $\omega_C/\omega_{LO} < 0.5$. In practice, measurements on bulk GaAs (Ref. 3) show that in this energy region $K_2(\text{tot}) = -1.75 \pm 0.1$.

In 2D systems there is universal agreement from a number of authors[9–11,26] that both the zero-frequency and the low-energy, frequency-dependent polaron enhancement of the effective mass are approximately three times as large as in bulk materials. The inclusion of screening and the finite wave function in the third dimension[10,12–15] both act to reduce the strength of the polaron coupling, although screening was not treated explicitly in magnetic fields. The two effects are interdependent and calculations using dynamic screening suggest that the polaron effects are reduced by a total factor of order 5.[13,14] Experimentally Sigg *et al.*[8] and Seidenbusch *et al.*[6] reached the qualitative conclusion that the polaron coupling in 2D was rather less than that seen in bulk GaAs. The results shown in Fig. 2 give a quantitative measure of this phenomenon. The energy dependence of the effective mass can be used to deduce a value of $K_2 = -1.4 \pm 0.1$, which is 20% less than that observed in the bulk material. This difference is approximately equal to the contribution to nonparabolicity from the polaron effects in the bulk, and would be consistent with a total screening of all polaron effects in the 2D electron gases studied here. No explicit theoretical treatments of screening in high magnetic fields have been reported, but the extreme degeneracy of the Landau levels in high quality samples may be expected to lead to very strong screening effects. The very narrow cyclotron resonance linewidths at low temperatures (of order 0.03 meV at 4.2 K) mean that the density of states within each Landau level is very much higher than without the field. Therefore the screening effects are very strong when the Fermi level lies within a Landau level and the statistics remain degenerate. This leads to a form of positive feedback in which increased screening reduces the scattering by charged centers, leading to a narrowing of the level widths and further enhanced screening, as has been discussed by Ando and Murayama.[27]

Direct evidence for the importance of screening and its dependence upon the density of states[28] comes from the oscillatory cyclotron resonance linewidths observed by Englert *et al.*[29] and also visible in the samples studied here.[17] Measurements have recently been reported by Rogers *et al.*[30] in which very low concentrations of electrons have been photoexcited into GaAs quantum wells and studied by cyclotron resonance. In this case there should be no significant screening, and a value of $K_2 = -2.3$ has been observed, which is consistent with a large unscreened 2D polaron contribution to the nonparabolicity.

All of the calculations discussed above assume that the electrons are interacting with bulk GaAs LO phonons. However, recent magnetophonon resonance experiments reported by Brummell *et al.*[31] on these samples show that the dominant optic-phonon absorption occurs at phonon frequencies significantly below the bulk GaAs LO-phonon value, suggesting that the electrons may be interacting with phonon modes associated with the presence of the interface. In the light of this result current theories must only be taken as a qualitative guide to the effects of polaron coupling in 2D systems.

IV. CARRIER CONCENTRATION DEPENDENCE OF m^*

When the carrier concentration changes in a heterojunction, the effective mass increases due to the nonparabolicity, with two separate energies contributing to the increase. Firstly, the additional carriers lead to an increase in the kinetic energy $\langle T \rangle_z$ due to the quantum confinement in the third (z) dimension. Secondly, the Fermi energy (E_F) increases, leading to an increase in the field necessary to achieve the quantum limit. The measurements of the carrier-concentration dependence of m^* were made in the low-field limit, which is that appropriate to all zero-field transport measurements. Thus we may write

$$\frac{1}{m^*} = \frac{1}{m_0^*}\left[1 + \frac{2K_2}{E_g}(E_F + \langle T \rangle_z)\right] . \tag{3}$$

Note that $\langle T \rangle_z$ is not necessarily equivalent to the subband energy E_i; for a triangular potential well $\langle T \rangle_z = E_i/3$,[32] and for a space-charge layer $\langle T \rangle_z$ for the lowest subband may be estimated from the variational wave function first proposed by Fang and Howard[33] for silicon inversion layers

$$\phi_0(z) = (\tfrac{1}{2}b^3)^{1/2} z e^{-bz/2} , \tag{4}$$

where $b^3 = 12 m^* e^2 (n_{\rm dep} + 11n/32)/\epsilon\epsilon_0 \hbar^2$ (Ref. 34) and $n_{\rm dep}$ and n are the depletion and inversion charge densities, respectively. The kinetic energy is then given by

$$\langle T \rangle_z = \frac{\hbar^2 b^2}{8m^*} . \tag{5}$$

The depletion charge densities were estimated by comparing the measured subband separations[17] with the calculations of Stern and Das Sarma[35] and were all found to be of order 3×10^{10} cm^{-2}. This is consistent with the doping levels of the p-type GaAs buffer layers ($N_A - N_D = 5\times10^{13}$ cm^{-3}), and gives values of $\langle T \rangle_z$ in the range 6–20 meV for n between 0.9×10^{11} and 9×10^{11} cm^{-2}.

Figure 4 shows the results of measurements of the carrier-concentration dependence of the effective mass $m^*(E_F)$, obtained by the use of a number of different samples. For each sample a range of carrier concentrations was studied by using the persistent photoconductivity. This procedure leads to some small deviations from the general curve, since measurements of the subband energies[17] have shown that the photoexcitation can act to reduce the depletion charge significantly, while increasing the inversion charge. Also shown in Fig. 4 is a fit to the carrier-concentration dependence using Eq. (3), with $m_0^* = 0.0663 m_e$ and $K_2 = -1.4$. The gradient of the m^* versus n curve is largely defined by K_2, while changes in m_0^* will move the entire graph vertically, and therefore the two variable parameters are largely independent, giving K_2 accurate to within ± 0.1. Approximately $\frac{1}{3}$ of this energy dependence is due to the variation of $\langle T \rangle_z$, and the rest to the change in E_F.

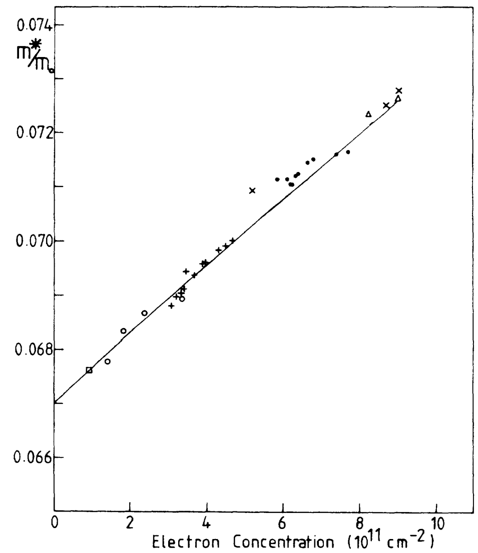

FIG. 4. The electron-concentration dependence of the cyclotron mass at 4.2 K, measured in the low-field limit where the mass is frequency independent. Several samples were studied, each represented by a different symbol, and the concentrations of each sample were varied using the persistent photoconductivity. The solid line is a fit to the points with $m_0^* = 0.0663 m_e$, $K_2 = -1.4$, and $n_{\rm dep} = 0.3\times10^{11}$ cm^{-2}.

This result is thus in good agreement with the change in m^* with energy found from the quantum limit measurements. More surprising perhaps is that the band-edge mass deduced ($0.0663 m_e$) is larger than found for bulk GaAs ($0.0660 m_e$) in similar measurements.[3] This would seem to contradict the conclusion from the nonparabolicity that the polaron interaction has been largely screened away, since this should lead to a lower band-edge mass. One possibility is that the penetration of the wave function into the $Ga_{1-x}Al_xAs$ barrier may influence the mass. The electron penetration is only of order 0.5%, but the evanescent form of the wave may result in an additional dressing of the electron mass. Further evidence for the change in the polaron enhancement of the mass is presented in the next section on the temperature dependence of m^*.

V. TEMPERATURE DEPENDENCE OF m^*

The temperature dependence of the effective mass has been measured at 119 μm in three samples (G63, G29, and G71) from 4.2 to 280 K. As the temperature increases, higher Landau levels will become thermally populated and since these higher-level transitions were not resolved they will lead to an increase in the measured effective mass due to nonparabolicity. The effective mass associated with transitions from the $L=0$ to $L=1$ Landau levels (m_{01}^*) has been calculated for each temperature by assuming that the measured mass represents an average over several transitions weighted by the populations of their initial levels, using the nonparabolicity of $K_2=-1.4$ deduced from the frequency and electron-concentration dependences of the mass. This correction to m^* is in fact small, being of order 0.7% at 100 K. The calculated values of m_{01}^* at an energy of 10.4 meV (119 μm) are plotted as a function of temperature in Fig. 5, with results from a sample of bulk GaAs also shown for comparison.

The unexpected feature of the 2D results is that m_{01}^* shows a rapid increase with temperature up to 100 K of approximately 2%, after which the mass falls slightly as in the bulk material. This behavior is attributed to a change in the polaron dressing of the effective mass caused by a change in the screening of the electron–optic-phonon interaction.[31] At low temperatures the Landau levels are highly degenerate, with a high density of states caused by the very narrow level widths which are reflected in the narrow cyclotron resonance linewidths. This will lead to a very strong screening of the polaron interaction, as discussed earlier, and hence there is almost no polaron dressing of the effective mass. As the temperature increases, the Landau levels will broaden [above 30 K the linewidths are limited by phonon scattering and vary approximately as $T^{3/2}$ (Ref. 36)] and electrons will begin to be distributed between different Landau levels. Both of these effects will lead to a reduction in the strength of the screening since both the density of states and the number of electrons within $k_B T$ of the Fermi level will fall, so that the polaron mass enhancement reappears. A similar argument has recently been used to explain apparent changes in the resonant polaron coupling in GaAs-$Ga_{1-x}Al_xAs$ heterojunctions when the electrons are heated by an electric field.[37] At a temperature of 120 K, $k_B T \simeq \hbar\omega_C$, and the electrons are distributed over several Landau levels. At temperatures above this, the screening should be weak and temperature independent,[29] consistent with the observed behavior of the effective mass shown in Fig. 5. An extrapolation of the high-temperature values of m_{01}^* to $T=0$, using the temperature dependence measured in bulk GaAs, will therefore give a direct measure of the polaron contribution to the effective mass at high temperatures, assuming that polaron effects can be completely ignored at low temperatures. This procedure gives an increase in mass of 1.8%, 2.1%, and 2.2% in samples G63, G29, and G71, respectively. These values can be considered as lower limits for the unscreened polaron mass enhancement in this quasi-2D system.

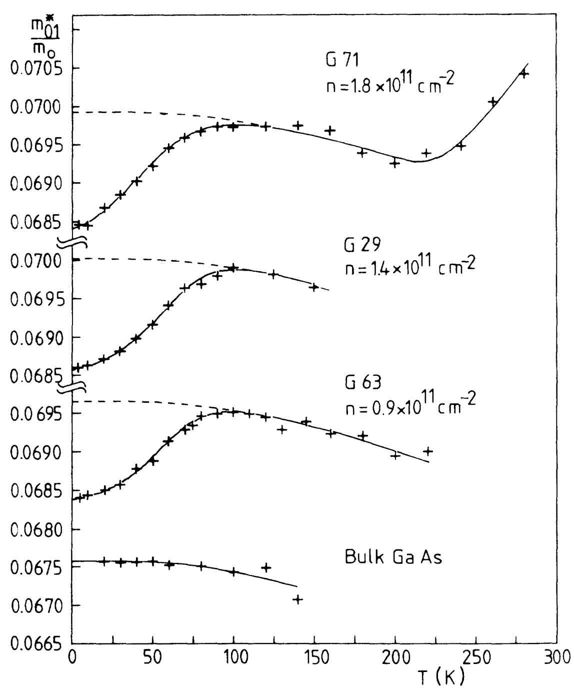

FIG. 5. The temperature dependence of m_{01}^*, the cyclotron mass due to the lowest transition, in samples G63, G29, and G71. For comparison, results from a sample of bulk GaAs are also shown. The heterojunctions show an unexpected fall in m_{01}^* below $\sim$100 K, which is attributed to strong screening of the polaron coupling at low temperatures. The increase in m_{01}^* above $\sim$220 K in sample G71 is believed to be due to the increasing influence of the $L=2$ to $L=3$ transition, which is strongly affected by resonant polaron effects. The dashed lines are extrapolations using the bulk dependence.

A theoretical polaron enhancement of 3.9% has been calculated for an ideal 2D system,[9,11] but this will be reduced by the finite extent of the wave function in the third dimension[10,12,15] and calculations by Das Sarma[12] give mass enhancements of 1.4%, 1.5%, and 1.6% for three samples, respectively. This discrepancy may be due to the correction procedure used to deduce m_{01}^* from the high-temperature resonances, in which the contribution from transitions starting at higher Landau levels was removed using the nonparabolicity deduced from the low-temperature measurements. However, we have just demonstrated that at low temperatures the electron-phonon interaction is screened, leading to a reduced nonparabolicity, but that it reappears at higher temperatures, which may cause an increased effective nonparabolicity. Thus the corrections could be larger than those which we have employed, leading to a reduction in the value deduced for m_{01}^* at higher temperatures. Using the calculated values for the magnitude of the polaron coupling[26] and correcting for the finite wave function in the third dimension[12] we can estimate a value for K_2 of -1.8, very close to that found in bulk GaAs where the polaron coupling is present to a similar extent. This then gives polaron enhancements for the three samples of 1.4%, 1.6%, and 1.7%, in excellent agreement with the predictions of theory and consistent with total screening of the polaron interaction at low temperatures. It is perhaps surprising that this agreement is so good in the light of the magnetophonon results on the same heterojunctions reported recently by Brummell *et al.*[31] It was found that the electrons appear to couple to phonons with an energy rather lower than that of the bulk GaAs LO phonon which has been assumed to dominate the polaron interaction in all the theoretical descriptions reported to date.

Finally, it may be noticed that in the data above 220 K, available only for sample G71, there is a rather more rapid rise in the effective mass with temperature. This is thought to be due to the increasing contribution of the $L=2$ to 3 Landau transition. The final state of this transition is close to the LO-phonon energy and hence will be strongly modified by resonant polaron coupling, leading to an increased effective mass. This has been studied in more detail by cyclotron emission work in both two[37] and three[1] dimensions.

VI. CONCLUSIONS

The total nonparabolicity in GaAs-$Ga_{1-x}Al_xAs$ heterojunctions has been deduced from the energy and carrier-concentration dependence of the effective mass and found to be 20% less than that measured in bulk GaAs. The parameter K_2 is found to be -1.4 ± 0.1, as compared with -1.75 for bulk GaAs in the same energy range. This difference is attributed, at least in part, to screening of the electron-phonon interaction. This conclusion is supported by the unexpected increase in m^*_{01} found as the temperature is raised from 4.2 to 120 K, which we believe to be due to the screening out of the polaron mass enhancement at low temperatures; as the temperature rises, level broadening and carrier redistribution reduce the screening, leading to the reappearance of the polaron mass enhancement.

The extent to which the polaron contribution to the nonparabolicity is screened is difficult to assess, given the uncertainties in calculating the exact magnitudes of both the interband and polaron contributions. However, the measured temperature dependence of m^* suggests that the nonresonant polaron mass enhancement is almost totally screened at low temperatures, which suggests that the polaron contribution to nonparabolicity will also be strongly screened. This is supported by the difference between the 2D and bulk nonparabolicities, which is close to the value calculated for the bulk polaron contribution. This argument implies that the band nonparabolicity in GaAs is underestimated by three-band $\mathbf{k}\cdot\mathbf{p}$ theories, a view supported both by recent experiments on bulk GaAs (Ref. 3), and by the calculations of Zawadzki and Pfeffer[23] and Braun and Rössler,[22] who find significantly higher nonparabolicities using five-band $\mathbf{k}\cdot\mathbf{p}$ theories.

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