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Electron effective mass in direct-band-gap GaAs$_{1-x}$P$_x$ alloys

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The electron effective mass of the conduction band in direct-band-gap GaAs$_{1-x}$P$_x$ alloys ($x < 0.4$) is reevaluated. A direct determination of m^* using the optically detected cyclotron resonance technique is presented for the low composition values. For higher x values the scattering times decrease because of alloying and it was not possible to carry out resonance experiments. Instead the diamagnetic shifts of the shallow-donor-to-acceptor recombination lines in magnetic fields up to 12 T were investigated. Within the framework of a simple perturbation approach the corresponding $m^*(x)$ values ($0.17 < x < 0.44$) could be deduced. The results are compared with a theoretical estimate based on the $\mathbf{k}\cdot\mathbf{p}$ theory. There is good agreement between theory and experiment, resulting in a new x dependence of the conduction-band mass in the direct-band-gap GaAs$_{1-x}$P$_x$ alloys: $m^* = 0.067 + (0.06 \pm 0.003)\,x$.

I. INTRODUCTION

The GaAs$_{1-x}$P$_x$ alloys have found applications in light-emitting diodes and detectors[1] for a long time, and as a result the physical properties of these alloys have been investigated in great detail. With this background, it is remarkable that almost no data are available on such a fundamental property as the electron effective mass in the direct-band-gap region ($x < 0.4$). Despite the fact that numerous experimental techniques for effective-mass determinations are available, the most frequently used values of m^* date back to those determined by the Faraday effect.[2] These values, quoted in standard reference tables today, seem surprisingly high. For example, for GaAs$_{0.8}$P$_{0.2}$ they found[2] $m^* = 0.11$ and for GaAs$_{0.7}$P$_{0.3}$, $m^* = 0.3$, which should be compared to the value in GaAs: $m^* = 0.067$. Furthermore, if the m^* values in the alloys, determined by the Faraday effect, are used to calculate the binding energies of, for instance, donors in GaAs$_{1-x}$P$_x$ in the effective-mass approximation, one gets values around 10 − 18 meV binding energies, which are much larger than those experimentally observed.[3]

The electron effective mass can be determined in different ways. An indirect way is to measure g^*, the conduction-electron, spin-splitting value, either on the donor electron-spin resonance (ESR) signal[4] or by a direct measurement of the electron-spin splitting in the conduction band, using the conduction electron-spin resonance (CESR) technique.[5] In the case of GaAs the small g^* value ($g^* = -0.44$) for the electrons makes a conventional ESR measurement of the donor resonances difficult (microwave frequency around 9 GHz) since the resonance usually falls outside the magnetic fields available in electromagnets. In the alloys the same situation seems to occur, even though it is normally very difficult to test since, often, only epitaxially grown materials are available. CESR, using the optical pumping techniques, has been applied successfully in GaAs and other III-V semiconductors.[5] However, this technique requires specially selected samples with a certain relationship between the radiative lifetime and the spin relaxation time. This requirement is not easily fulfilled, and very special samples have to be found, making the technique, in most cases, impractical.

Conventional cyclotron resonance (CR) measurements make it possible to determine m^* directly. The potential of this technique was beautifully demonstrated in the first experiments in Si and Ge.[6,7] The CR technique requires doping (or light illumination) in order to produce free carriers and large single-particle scattering times τ_s in order to fulfill the cyclotron resonance condition $\omega_c \tau_s > 1$, where ω_c is the cyclotron frequency. The latter criterion for the observation of cyclotron resonance is not easily met in GaAs$_{1-x}$P$_x$ since the single-particle relaxation time decreases rapidly for $x > 0.2$ due to alloy scattering.[8] If, instead, the optically detected cyclotron resonance (ODCR) technique is used, some of the disadvantages connected with the conventional cyclotron resonance technique can be avoided.

In the ODCR technique one detects cyclotron resonance on the photoluminescence signal caused by exciton or donor-acceptor recombinations.[9,10] The mechanism is that free carriers are effectively heated by microwaves or far-infrared (FIR) photons at CR conditions. The hot electrons can either impact-ionize the bound excitons, or electrons, or heat the lattice.[11,12] In both cases the effect on the PL signals is strong, and the CR condition can therefore be monitored, in a very sensitive way, directly by the PL intensity. Besides the fact that the method requires no doping, and that thus no band-filling effects

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are observed, it is well documented that the technique reduces the electron scattering by ionized impurities by optically neutralizing the impurities. Increased single-particle relaxation times (more than a factor of 10 are reported[10]) lead to better $\omega_c\tau_s$ values, and thereby higher sensitivities. It should be noted, however, that alloy scattering is a severe problem also in the ODCR technique.

If the above-mentioned techniques fail, the shift of the exciton, or donor-acceptor pair, recombination lines with magnetic field (diamagnetic shift) can be used to determine m^*.[13,14] This technique is fairly insensitive to the sample quality as long as a luminescence signal is observed. It is, however, an indirect technique and requires knowledge of the physical origin of the luminescence (free exciton, bound exciton, donor-acceptor pair, etc.) and a theory[15,17] describing the relevant recombination process in an applied magnetic field.

In this paper we report on measurements of m^* using the ODCR technique and the analysis of the diamagnetic shift in magnetic fields up to 12 T of the shallow donor-acceptor recombination in $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ alloys up to the crossover from the direct to the indirect gap at $x = 0.45$. The results are compared with a five-band $\mathbf{k}\cdot\mathbf{p}$ calculation in which the x dependence of the lowest- and higher-lying conduction bands are taken into account. The result is a dependence of m^* on x for direct-band-gap $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ alloys.

II. EXPERIMENT

The $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ samples were grown at atmospheric pressure in a metal-organic vapor-phase epitaxy (MOVPE) reactor. They were grown on both semi-insulating and n^+-conducting substrates and contain a 1-μm buffer layer followed by a strain relieving, graded layer (0 – 10 μm, depending on the final composition) and, finally a 5-μm alloy with the desired composition. The background carrier concentration was $5 \times 10^{15}\,\mathrm{cm}^{-3}$ – $1 \times 10^{16}\,\mathrm{cm}^{-3}$, with the high value for the P-rich material. For the determination of the composition we used the position of the principal exciton luminescence line at 4 K. The exciton line position has previously been correlated with an x-ray analysis using a microprobe in a scanning electron microscope.

The experimental setup is shown schematically in Fig. 1. The photoluminescence (PL) measurements were performed using either a HeNe laser (632.8 nm, 3 mW) or an Ar ion laser (514.5 nm, 20 mW). The laser beam was focused by a camera lens onto a glass fiber which ended directly on the front side of the sample, which in turn was placed at the center of a 12-T solenoid magnet located in a He cryostat (Cryogenic Consultant). The luminescence light from the sample was collected by the same fiber and, using the same camera lens, focused on the entrance slit of a single, 0.22-m focal-length monochromator. The spectral resolved light (2-nm resolution between 500 – 900 nm) was detected by a LN_2-cooled Ge detector (North-Coast). The exciting laser light was mechanically chopped (< 1 kHz) in order to make lock-in detection possible.

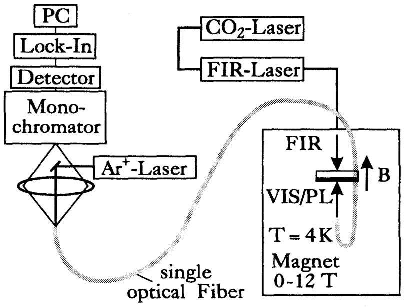

FIG. 1. Sketch of the experimental setup for ODCR ($\lambda_{\mathrm{FIR}} = 118.8\,\mu$m) and magnetoluminescence in the Faraday configuration.

In the ODCR measurements the sample was simultaneously irradiated from the back (semi-insulating GaAs was used as substrate material) by a CO_2-pumped, far-infrared laser (Edinburgh Instruments) working at 118.83 μm. The FIR light was guided by mirrors to the sample and was amplitude-modulated by a mechanical chopper. The FIR power was held at 10 mW to avoid heating effects in the sample. The sample temperature was < 6 K in all experiments.

III. RESULTS

A. ODCR measurements

The band mass m^* can be determined with high precision using cyclotron resonance if the single-particle scattering time and the frequency used are such that $\omega_c\tau > 1$. m^* can then be calculated directly from B_{res}, the position of the resonance peak. The only uncertainty stems from the magnetic-field position, an uncertainty which is usually smaller than 1% with calibrated Hall sensors. In this work we used the ODCR technique, which was briefly described in the Introduction. In order to observe well-resolved, high-intensity CR lines it is advantageous to work at high frequencies, i.e., in the FIR range (70 – 380 μm). The experimental ODCR data were obtained on the donor-acceptor (DA) recombinations. Before going into detail we will briefly describe the photoluminescence results on which the ODCR detection is based.

In Fig. 2 we show a typical photoluminescence spectrum for $x = 0.31$. The two well-resolved peaks arise from excitonic and donor-acceptor recombinations with an energy separation of 26 meV. Carbon has a binding energy of 27 meV in GaAs (Ref. 18) and is the dominant acceptor in MOVPE-grown GaAsP samples. We therefore assume that the DA recombination involves carbon as the acceptor.

The DA nature of the low-energy band is further substantiated by investigating the behavior under low- and high-power excitation with the laser. In Fig. 2(b) we note that the transition moves to lower energies when the power is reduced by a factor of 100, whereas the excitonic lines are unshifted. Distant DA pairs, which are favored by low-power excitation, have a reduced Coulomb interaction due to the charged donors and acceptors and hence emit at lower energies.

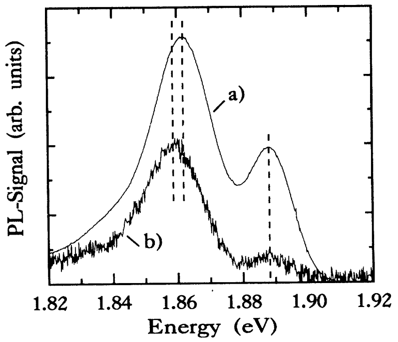

FIG. 2. The PL of $GaAs_{0.69}P_{0.31}$ at (a) high and (b) low excitation power reveals the DA nature of the low-energy band.

We summarize our PL experiments in Fig. 3. The sequence exciton and DA recombination were observed for all samples investigated. The peak positions of the excitons follow the Γ band, and our results are in line with previous works.[3,19] For comparison the dependence of the X conduction band on composition is also shown. One notes the crossover from direct to indirect band gap at $x = 0.45$.

In the ODCR investigations we monitored the DA recombination line, which was the strongest in all samples. The changes induced on the photoluminescence by the CR when using 118.83-μm FIR photons are shown in Fig. 4 for $x = 0.05$ and $x = 0.17$. We note that for the higher x value the resonance line is broader, reflecting the severe decrease in the scattering times with alloying. Indeed, samples with higher phosphorus compositions showed hardly any ODCR signals. The dependence on the electron mobility at room temperature of the composition[8] shows a steep decrease over one order of magnitude for x values above 30%. The m^* values determined from the B_{res} positions are collected in Fig. 5.

B. Diamagnetic shift

To extend our determination of the electron mass to values above $x = 0.17$, we used the well-established concept of studying the shift of the PL signals in magnetic

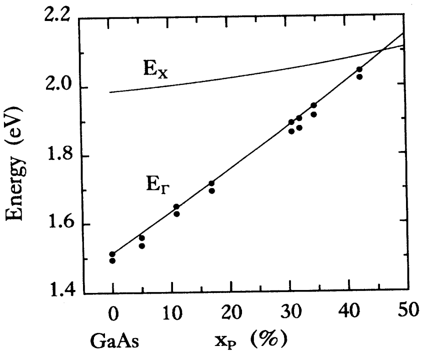

FIG. 3. The PL bands of the $GaAs_{1-x}P_x$ samples follow the direct-band-gap energy E_Γ.

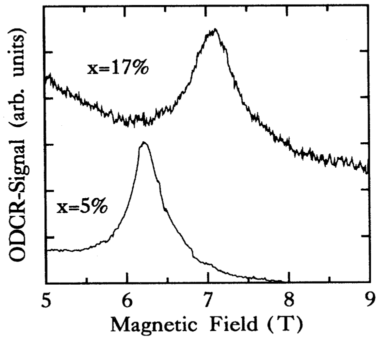

FIG. 4. The ODCR of the samples shows clear resonances allowing a precise determination of m^*.

fields from $B = 0\,\mathrm{T}$ to $B = 12\,\mathrm{T}$ (diamagnetic shifts).[13,14] Such dependence of the donor-acceptor recombination on magnetic field shows nonlinear shifts (proportional to B^2) of the peak maxima (see Fig. 6). The shift is about 3 meV for $x = 0.35$. The dashed line in Fig. 6 serves only as a guide for the eye.

The diamagnetic shift is proportional to $(a_0 B)^2$, where a_0 is the Bohr radius of the defect under consideration. In our case the contribution from the shallow acceptor is too small to be resolved and can therefore be neglected. Consequently, the diamagnetic shifts reflect the properties of the donors only. For the very small binding energies of the shallow donors, in the order of 6 – 8 meV, we can assume that there are no differences in the effective mass of the electrons located on the shallow donors and those located in the conduction band (see below). In the hydrogenic model,[15] using first-order perturbation theory, the diamagnetic shift is given by

$$\Delta E = \tfrac{1}{2} R^* \gamma^2, \tag{1}$$

with $\gamma = \hbar\omega_c / 2R^*$, where $\hbar\omega_c$ is the cyclotron resonance energy and $R^* = m^* e^4 / 2\hbar^2 \epsilon^2$ ($R^* = R_0 m^* / \epsilon^2$, where R_0 is the Rydberg constant), m^* is the effective electron mass in units of m_0, and ϵ is the static

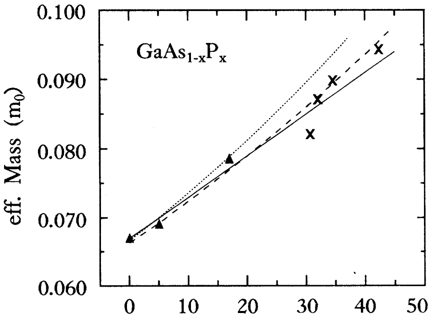

FIG. 5. Values for m^* in $GaAs_{1-x}P_x$. Triangles mark ODCR and crosses indicate magnetoluminescence results. The solid line is the best linear fit $m^* = 0.067{+}0.06(\pm 0.003)x$. The results of the $\mathbf{k}\cdot\mathbf{p}$ theory with constant (dotted line) and varying matrix elements (dashed line) are also given.

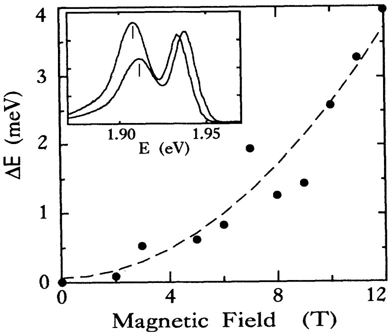

FIG. 6. The magnetoluminescence for $B = 0\,\mathrm{T}$ and $B = 12\,\mathrm{T}$ is shown in the inset ($x = 0.35$); the shift of the DA line behaves diamagnetic as sketched by the dashed parabola.

dielectric function. With $\omega_c = eB/m^*$ one obtains $\Delta E = 1.23 \times 10^{-10}\, B^2\epsilon^2/m^{*3}\,(\mathrm{eV/T^2})$. Taking a linear interpolation for $\epsilon = 12.53 - 1.65\,x$ in the $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ alloys,[16] and applying $B = 12\,\mathrm{T}$, Eq. (1) can be rewritten as

$$\Delta E = 1.77 \times 10^{-8} \frac{(12.53 - 1.65\,x)^2}{m^{*3}}\ \mathrm{eV}. \tag{2}$$

Using Eq. (2) and the experimentally determined diamagnetic shifts (ΔE), the $m^*(x)$ values for the different compositions above $x = 0.2$ were deduced (see crosses in Fig. 5). A best linear fit to the experimental data (solid line in Fig. 5) gives $m^*(x) = 0.067 + (0.06 \pm 0.003)x$.

For low alloy compositions ($x < 0.2$) the diamagnetic energy shifts are comparable to the binding energy of the donor electron, and the perturbation approach can be questioned. For higher alloy compositions the approximation is substantially better, even though not perfect. We will, therefore, compare the results based on the perturbation theory with those obtained with the variational approach used by Aldrich and Greene,[17] a method which is valid for a hydrogenlike system in an arbitrary magnetic field. We note that for $x = 0$ the two theories give identical results only for very low magnetic-field values. For 12 T the perturbation theory clearly overestimates the shift. For $x = 0.4$ and $m^* = 0.095$ both theories give identical results even up to 12 T. The reason is that the diamagnetic shift is smaller, and the binding energy is slightly larger for higher x values. For intermediate x values the perturbation approach gives results that are slightly wrong. However, m^* enters as the third power [see Eq. (2)], which means that even a difference of a factor of 2 in diamagnetic shift gives only a 20% decrease of the electron mass. Therefore, in the determination of m^* using the diamagnetic shift up to 12 T, perturbation theory is applicable even for $x > 0.2$.

It should be mentioned that for higher phosphorus concentrations the shifts become smaller and the linewidths of the emission lines increase due to alloy broadening, making a determination of the peak position less accurate. In spite of this we feel confident that the overall trend in $m^*(x)$ is correct.

In the following we support the results by an estimate of m^* using the five-band **k·p** theory.

TABLE I. Relevant band-structure parameters in GaAs (Ref. 23).

$E_0 = 1.519\,\mathrm{eV}$	$E(\Gamma_8^c) - E(\Gamma_8^v) = 4.659\,\mathrm{eV}$	$p^2 = 28.9\,\mathrm{eV}$
$\Delta_0 = 0.341\,\mathrm{eV}$	$E(\Gamma_7^c) - E(\Gamma_8^v) = 4.49\,\mathrm{eV}$	$p'^2 = 6\,\mathrm{eV}$
		$C = -2$

C. Theoretical estimate of m^* using five-band k·p theory

In their classical work on the **k·p** theory in III-V compounds and alloys Hermann and Weisbuch[23] derived expressions for m^* which first took into account the precise nature of the band structure of the semiconductor and, second, related m^* to the interband matrix elements p^2, which couple conduction bands and valence bands. Explicitly m^* is given by

$$\frac{m_0}{m^*} = \frac{p^2}{3}\left[\frac{2}{E_0} + \frac{1}{E_0 + \Delta_0}\right] - \frac{p'^2}{3}\left[\frac{2}{E(\Gamma_8^c) - E_0} + \frac{1}{E(\Gamma_7^c) - E_0}\right] + C. \tag{3}$$

For GaAs all relevant parameters, as presented in Ref. 23, are given in Table I. For notations, see Ref. 23. In the direct-band-gap $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ alloys we first assume that the interband matrix elements p^2 and p'^2 do not change. For the x dependence of E_0 we use the 4-K data:[20] $E_0(x) = 1.519 + 1.712x + 0.186x^2$ (eV). For the higher conduction bands only room-temperature values are available:[20–22] $E\{\Gamma_8^c\}(x) = 4.659 + 0.14x$ (eV) and $E\{\Gamma_7^c\}(x) = 4.49 + 0.25x$ (eV). These values are sufficient for our purpose since the second term in Eq. (3), proportional to p'^2, gives only a 10% correction to the calculated mass. C is approximated to be constant with x.

The calculated m^* values for the investigated alloy composition range are plotted in Fig. 5 (dotted line). We note that the agreement with the mass dependence obtained from the CR experiments and the diamagnetic shift (triangles and crosses) is satisfactory. The agreement can be improved under the quite reasonable assumption that p^2 is slightly dependent on x. The best fit obtained includes such a variation with $p^2 = 28.9 + 2.5x$ (eV) (dashed line).

IV. DISCUSSION

The reliability using the diamagnetic shift for an m^* determination is based on the precise nature and description of the luminescence transition investigated. The neutral donor is for the analysis the most simple defect (e.g., the donor taking part in the donor-acceptor recombination, when the acceptor does not contribute to the diamagnetic shift). Comparing the donor binding energy E_B with the diamagnetic shift ΔE, the perturbation approach should be valid as long as $\Delta E \ll E_B$. For $0 < x < 0.2$ this is only the case for small magnetic fields. In this composition range E_B increases from 5 meV to 7.2 meV. For $0.2 < x < 0.4$ E_B increases further up to

8.7 meV. This should be compared to the experimentally observed ΔE of 2.8 meV at 12 T for $x = 0.4$.

In some cases the perturbation theory also holds for neutral, acceptor-bound excitons. In the "pseudodonor" model one assumes that the two holes are strongly bound by the short-range potential of the acceptor. The additional positive charge produces a long-range Coulomb potential, which binds the electron in a donorlike state. In those cases the analysis of the diamagnetic shifts resulted in m^* values very close to the free-electron values (deviations $< 3\,\%$).

Although the error between the experimental and calculated diamagnetic shift introduced by the use of first-order perturbation theory might, for $x > 0.2$ and for 12 T, be as large as 8 %, it seems not to influence the overall trend in the x dependence of m^* (see discussion above). This is nicely confirmed by the ODCR measurements for $x < 0.2$. Instead, the limitation in the diamagnetic shift measurements is mainly the low spectral resolution in the detection system. This low resolution is, however, beneficial in the ODCR measurements since a high intensity (obtained by low-resolution measurements) is needed in order to detect the small changes in the PL signal upon FIR irradiation.

The theoretical approach based on the five-band $\mathbf{k}\cdot\mathbf{p}$ method[23] gives a clear trend in the m^* values, immediately ruling out the extremely high values of m^* at $x = 0.2$ and $x = 0.3$ as deduced in the Faraday effect investigation.[2] The precision of the $\mathbf{k}\cdot\mathbf{p}$ method in the case of $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ is, however, not better than 10%, probably because of the assumption of a "constant" C and the contribution from p'^2. Clearly, investigations where both m^* and g^* (which is determined almost entirely by p^2) could be measured directly in the same samples would be desirable. That would make possible the estimation of both p^2 and p'^2 as a function of the alloy composition.

Probably the best, and most precise, method to measure m^* in the alloys is cyclotron resonance. Besides sharing all common advantages with CR, the increased sensitivity of the optical detection method (quantum transformation of FIR quanta to optical quanta) makes this method even more powerful in many cases. Limitations due to alloy scattering are an inherent problem of the alloy system, and apply to all CR techniques, also ODCR. Improvements might be possible either by pulsed/delayed techniques for the FIR laser or photocounting systems in the PL detection.

V. CONCLUSIONS

The electron effective mass of the lowest Γ_5 conduction band in direct-band-gap $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ alloys ($x < 0.4$) has been studied using cyclotron resonance techniques and by measuring the shifts of the DA recombination lines in magnetic fields up to 12 T. This diamagnetic shift is analyzed within the framework of a simple perturbation approach. The resulting $m^*(x)$ values are compared with a theoretical estimate based on the $\mathbf{k}\cdot\mathbf{p}$ theory. There is good agreement between theory and experiments. The result is an x dependence of the Γ_6 conduction-band mass in the direct-band-gap $\mathrm{GaAs}_{1-x}\mathrm{P}_x$ alloys: $m^* = 0.067 + (0.06 \pm 0.003)\,x$.

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