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Split-Off Valence-Band Parameters for GaAs from Stress-Modulated Magnetoreflectivity

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We have measured the spin-orbit split-off valence-band parameters in high-purity epitaxial GaAs at ~30 °K by means of stress-modulated interband magnetoreflectivity. Our results are $(m/m_c - m/m_{so}) = 21.5 \pm 0.4$, $g_c + g_{so} = -4.7 \pm 1.0$, where m_c and m_{so} are the conduction-band and split-off-band effective masses and g_c and g_{so} are the corresponding effective g factors. From these results, we deduce $m_{so}/m = -0.154 \pm 0.010$, $g_{so} = -4.9 \pm 1.0$, and $\kappa^L = 1.2 \pm 0.25$, where κ^L is related to the antisymmetric constant introduced by Luttinger.

I. INTRODUCTION

At the center of the Brillouin zone, the spin-orbit split-off valence band in GaAs lies at an energy $\Delta \approx 0.34$ eV below the degenerate light- and heavy-hole bands. Optical transitions from this band to the conduction band have previously been observed in both absorption[1] and electroreflectance.[2] In this paper, we present the first experimental results of magneto-optical effects associated with this transition. From our data, we are able to determine the first experimental values for the effective mass and the effective g factor for the split-off band. We used the differential technique of stress modulation[3,4] together with samples of epitaxially grown n-type GaAs prepared by Lincoln Laboratory. Stress-modulated or electric-field-modulated magnetoreflectivity has previously provided data on the split-off bands in Ge,[5,6] InSb,[7] InAs,[8] and GaSb.[9] The extension of this type of experiment to GaAs, however, was not possible until the epitaxial process made available material of exceptional purity.

The first estimate of Δ, the spin-orbit splitting, for GaAs was made by Braunstein[10] from inter-valence-band-absorption measurements in p-type material. He found $\Delta \approx 0.33$ eV; using this together with Kane's expressions,[11] he estimated the ratios of the split-off-band effective mass to those of the light- and heavy-hole bands. In the same way, Ehrenrich[12] deduced the value $m_{so} \approx -0.2m$ for the split-off-band effective mass. Sturge[1] measured the absorption edge for transitions from the split-off band to the conduction band and found $\Delta = 0.35 \pm 0.01$ eV. Seraphin[2] studied this transition with the electroreflectance technique as a function of temperature from 200 °K to 375 °K. He determined $\Delta = 0.348 \pm 0.002$ eV and showed that the temperature dependence of Δ was negligible compared to that of the energy gap. In later electroreflectance experiments on GaAs -

GaP alloys at room temperature, Thompson *et al.*[13] obtained the value $\Delta = 0.339 \pm 0.003$ eV, and Williams and Rehn[14] obtained $\Delta = 0.340 \pm 0.004$ eV. Recently, Nishino *et al.*[15] determined the value 0.350 ± 0.004 eV in electroreflectance studies over a temperature range between 300 and 25 °K.

In the technique of stress modulation[4,5] a thin (~0.01-in.) sample is bonded with vacuum grease to a piezoelectric transducer. The transducer is mounted on the cold finger of a liquid-helium Dewar. We estimate that the vacuum grease forms a rigid bond below ~ 160 °K, so that the sample is strained after cooling to its final temperature of ~ 30 °K. The strain is small, resulting in shifts of the band edges by a few meV which can be taken into account with the analysis of Kleiner and Roth.[16] We used a glass Fresnel rhomb to produce circularly polarized light, and a silicon photodiode to detect the reflected radiation. The sample orientation was such that the [211] direction was normal to the transducer surface and parallel to the direction of the magnetic field. The other details of the experimental apparatus are given in the preceding paper.[5] The are of the reflecting surface of the sample was $\frac{3}{8} \times \frac{3}{16}$ in. The sample temperature was ~ 30 °K, at which the electron concentration was ~ 8×10^{13} cm^{-3} and the electron mobility was ~ 10^5 cm^2/V sec. The donor and acceptor concentrations were $N_D \approx 2 \times 10^4$ cm^{-3}.

The conduction and split-off bands in GaAs near $\vec{k} = 0$ are spherical. Furthermore, interband magnetoabsorption experiments by Vrehen[17] show that the nonparabolicity of the conduction band is relatively small. Consequently, we chose to interpret our data in terms of the simplest model possible. We assume that the Landau levels of the conduction band are given by

$$\mathcal{E}_n^c = \mathcal{E}_g + \delta\mathcal{E}_g + s\left[\left(n + \frac{1}{2}\right)\frac{m}{m_c} + \frac{1}{2} g_c M_S\right] ,$$

where $s = \hbar eH/m_c$, with H the magnetic field and m the free-electron mass, and where $M_S = \pm\frac{1}{2}$. Here n is the Landau quantum number, and $\delta\mathcal{E}_g$ is the strain-induced shift of the conduction band relative to the split-off band. Similarly, the levels of the split-off band are given by

$$\mathcal{E}_{n'}^{so} = -\Delta + s\left[\left(n' + \frac{1}{2}\right)\frac{m}{m_{so}} + \frac{1}{2} g_{so} M_J\right] .$$

Allowed transitions occur between these levels subject to the selection rules $n - n' = 0$, and $M_S - M_J = +1$ for LCP and $M_S - M_J = -1$ for RCP. Here LCP and RCP denote left and right circularly polarized light propagating in the direction of the magnetic field. The allowed transition energies are

$$\mathcal{E}_n = \mathcal{E}_n^c - \mathcal{E}_n^{so} - \Delta_{ex} = \mathcal{E}_g + \delta\mathcal{E}_g + \Delta - \Delta_{ex} + s\left[\left(n + \frac{1}{2}\right)\times\left(\frac{m}{m_c} - \frac{m}{m_{so}}\right) \pm \frac{1}{4}(g_c + g_{so})\right], \qquad (1)$$

with the plus sign for LCP and the minus for RCP. Here we have included the exciton binding energy Δ_{ex} associated with split-off-band-to-conduction-band transitions.

In the next section we compare our experimental data to Eq. (1). In Sec. III we draw upon the theoretical analysis of Pidgeon and Brown,[18] in which the conduction, light- and heavy-hole, and split-off bands are coupled, to show more clearly the interrelation between the split-off-band parameters and those of the other three bands.

II. EXPERIMENTAL RESULTS

In Fig. 1, we plot the experimentally determined quantity $(1/R)(\Delta R/\Delta S)$ as a function of photon energy for $H = 0$ and $H = 88.6$ kG. Here R is the reflectivity and ΔR is the change in R due to a change ΔS in the strain in the plane of the sample. Each curve shown represents a point-by-point average by computer of four experimental traces. The structure in the spectrum for $H = 0$ represents the onset of split-off-band-to-conduction-band transitions. Similarly, the structure in the spectrum for $H = 88.6$ kG represents the thresholds for allowed transitions between successive pairs of Landau levels. The LCP spectrum was essentially the same as the corresponding RCP spectrum, as indicated by that portion of the LCP spectrum shown in Fig. 1, except for a displacement to lower energy by 2.4 ± 0.5 meV. This energy difference can be interpreted in terms of Eq. (1) to give a value for the sum of the effective g factors:

$$g_c + g_{so} = -4.7 \pm 1.0. \qquad (2)$$

The conduction-band g factor in GaAs is small; Roth's expression,[19] Eq. (13), gives $g_c \approx 0.2$. Using this value we obtain $g_{so} = -4.9 \pm 1.0$ from Eq. (2).

In order to further analyze the data of Fig. 1 it is necessary to make an assumption as to what particular features of the experimental spectra correspond to the transition energies of Eq. (1). Mavroides[20] has given the following empirical criterion for interpreting the line shapes obtained with stress modulation: The transition energies $\mathcal{E}_n$ are to be read at the minima of the $(1/R)(\Delta R/\Delta S)$ spectrum for $H > 0$, and at the inflection point for zero magnetic field. We have applied this criterion to the

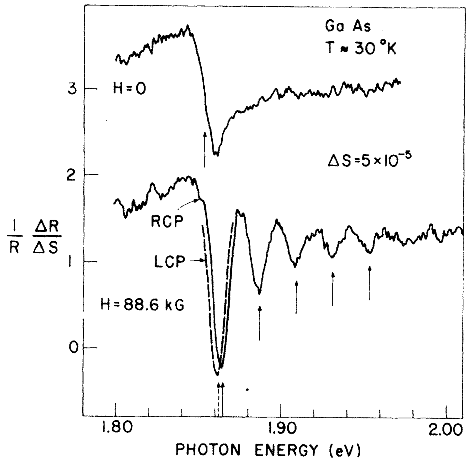

FIG. 1. Stress-modulated reflectivity spectra for $H=0$ and $H=88.6$ kG. $(\Delta R/R)$ is the relative change in reflectivity for a change ΔS in the strain in the plane of the sample. The $H=0$ spectrum has been displaced upwards for clarity. Arrows indicate the theoretical positions of the transition energies.

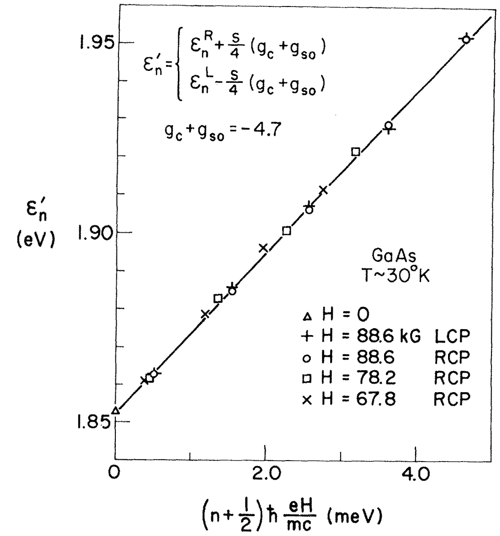

FIG. 3. Points are the experimentally observed minima in the $(1/R)$ $(\Delta R/\Delta S)$ spectra, taken from Fig. 2 for the higher values of H and for various values of n, and plotted against $(n+\frac{1}{2})s$, where $s=\hbar eH/mc$. The points were adjusted to eliminate the difference between the LCP and the RCP data. The solid line is a least-squares fit to the data.

data shown in Fig. 1, as well as to spectra taken for several lower values of H, and have plotted the transition energies so obtained as a function of H in Fig. 2. The solid lines are plots of Eq. (1) with $(g_c+g_{so})=-4.7$ and with a value of $(m_c^{-1}-m_{so}^{-1})$ determined by the following procedure: The points in Fig. 2 for higher values of H, for which the minima in the spectra were better resolved, were corrected for spin splitting, i.e., the quantity $\frac{1}{4}(g_c+g_{so})s$ was subtracted from the LCP points and added to the RCP points. These corrected points were then plotted against the term $(n+\frac{1}{2})s$ as shown in Fig. 3. Equation (1) predicts a straight line and this is obtained. The slope of this line gives

$$m/m_c - m/m_{so} = 21.5 \pm 0.4. \tag{3}$$

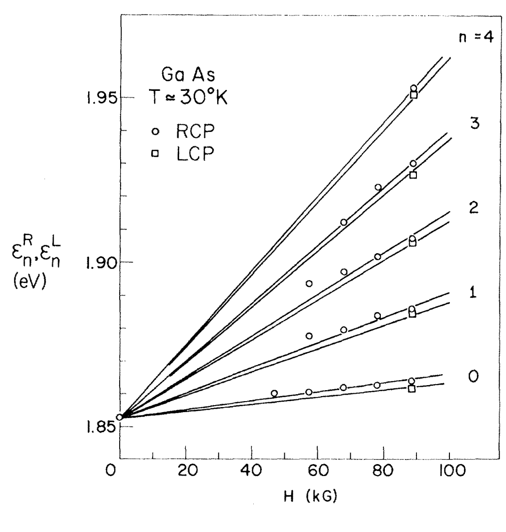

FIG. 2. Points are experimentally observed minima in the $(1/R)$ $(\Delta R/\Delta S)$ spectra for several values of magnetic field H. The Landau quantum number n labels the transitions. Solid lines are plots of Eq. (1) for $(g_c+g_{so})=-4.7$ and $(m_c^{-1}-m_{so}^{-1})=21.5m^{-1}$.

Stillman *et al.*[21] have recently done experiments on the magnetospectroscopy of shallow donor states using epitaxial GaAs samples similar to ours. They found $m_c=(0.0665\pm0.0005)m$, which is in agreement with both Vrehen's magnetoabsorption value[17] of $(0.067\pm0.002)m$ and the recent cyclotron resonance value of $(0.0648\pm0.0015)m$ obtained by Chamberlain and Stradling.[22] Using Stillman's value, we obtain from Eq. (3) $m_{so}=-(0.154\pm0.010)m$.

The experimental point for $H=0$ in Fig. 3 corresponds to the position of the arrow in the upper trace of Fig. 1, i.e., to the inflection point. The minimum of the $H=0$ trace is about 6 meV higher. The fact that the best fit to the magnetic field data of Fig. 3 passes through the inflection point at $H=0$ and not through the minimum shows that our data are at least consistent with the criterion given above. The intercept at $H=0$ of the least-squares fit in Fig. 3 gives, according to Eq. (1),

$$\mathcal{E}_g + \delta\mathcal{E}_g + \Delta - \Delta_{ex} = 1.8524 \pm 0.0005 \text{ eV}. \tag{4}$$

An estimate for Δ can be obtained from Eq. (4) together with our data for the stress-modulated reflectivity spectrum for the fundamental edge. These data, shown in Fig. 4, were taken under exactly the same conditions as were the previous data. This means that the stress-induced energy-gap shift $\delta\mathcal{E}_g$ is the same for both sets of data. The spectrum of Fig. 4 exhibits a broad negative peak at ~ 1.490 eV, a sharp (2 – 3-meV-wide) negative peak at 1.511 eV, and possibly a weak negative peak at 1.514 eV. The broad minimum at ~1.490 eV is no doubt due to an impurity transition. Several authors[2,14,23] have reported strong structure in the electroreflectance spectrum of n-type GaAs near this energy, about 30 – 35 meV below the energy gap.

The sharp negative peak at 1.511 eV is more difficult to interpret. Sturge[1] studied the absorption edge of high-resistivity GaAs and determined the values 1.521 and 1.518 eV (each ± 1.5 meV) for the energy gap at 21 and 55 °K, respectively. The sample temperature for the present experiment was ~ 30°K, for which the value 1.520 ± 0.002 eV is a good estimate; this value is indicated by the arrow in Fig. 4. Sturge obtained a value of 3.3 – 3.4 meV for the free-exciton binding energy $\mathcal{E}_{ex}$. Recent high-resolution low-temperature photoluminescence experiments[24] in epitaxial GaAs, however, have determined the binding energy $\mathcal{E}_{ex}$ to be 4.7 ± 0.4 meV. For this latter value, the free-exciton transition would occur at 1.515 eV, which would be above the energies of both the sharp negative peak and the weak minimum just above it. A possible interpretation for this structure is that it represents the two components of the free-exciton transition which have been split apart and shifted to lower energy by the dc strain.[16] This is unlikely in view of the order-of-magnitude difference in the intensities of the two negative peaks.

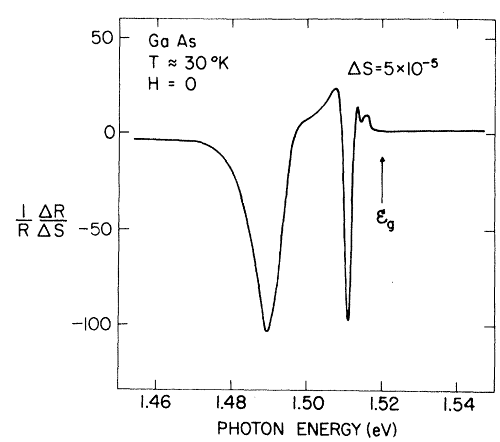

FIG. 4. Stress-modulated reflectivity spectrum $(1/R)(\Delta R/\Delta S)$ at the fundamental edge of GaAs. A smooth curve was drawn through the data points.

A more probable interpretation for the sharp negative peak at 1.511 eV is that it is associated with transitions involving a bound-exciton-impurity complex. High-resolution photoreflectance experiments on n-type epitaxial GaAs by Shay and Nahory[25] have shown that at 2 °K the dominant feature of the laser-modulated $(\Delta R/R)$ spectrum is a sharp (~3 meV) wide negative peak about 0.5 meV below the free-exciton transition energy. They attributed this structure, which is very similar to that of Fig. 4, to a bound-exciton transition. This interpretation for the sharp negative peak in Fig. 4 leaves some uncertainty as to the magnitude of the dc strain in the sample since the deformation potentials for impurity levels are generally different from the known potentials for the band edges. If both this difference and the difference between the binding energies for bound and free-exciton transitions are neglected, the energy of the sharp minimum in Fig. 4 can be set equal to

$$\mathcal{E}_g + \delta\mathcal{E}_g - \mathcal{E}_{ex} = 1.511 \text{ eV}. \qquad (5)$$

This equation can be combined with Eq. (4) to give an equation involving Δ, Δ_{ex}, and $\mathcal{E}_{ex}$. The binding energy Δ_{ex} can be estimated from the simple hydrogenic formula

$$\Delta_{ex} = (\mu/m)\, R_0/\epsilon^2 \,, \qquad (6)$$

where R_0 = 13.62 eV. The inverse reduced mass ratio (m/μ) is given in Eq. (3) and the static dielectric constant ϵ has been measured[26] as 12.53 ± 0.10. For these values, Eq. (6) gives Δ_{ex} = 4.0 meV. The resulting estimate for the spin-orbit splitting Δ is 0.341 eV, which is close to the values of Thompson *et al.*[13] and of Williams and Rehn.[14]

III. DISCUSSION

The parameters m_{so} and g_{so} are not unrelated to the analogous parameters characterizing the conduction and light- and heavy-hole bands. This is evident, for example, in the theoretical analysis of Pidgeon and Brown,[18] in which the energy levels of these four coupled bands are calculated in a magnetic field. From their formulation, one can obtain the following expressions for the parabolic region of the split-off band edge[5]:

$$m/m_{so} = -\gamma_1^L + (E_p/3\mathcal{E}_g)[\Delta/(\mathcal{E}_g+\Delta)] \,, \qquad (7)$$

$$g_{so} = -2\{2\kappa^L + 1 - (E_p/3\mathcal{E}_g)[\Delta/(\mathcal{E}_g+\Delta)]\} \,. \qquad (8)$$

TABLE I. The first five columns list the results of other experiments for the light- and heavy-hole band parameters. The last two columns list the split-off band parameters predicted in each case by Eqs. (7) and (11).

	$-m_l/m_h$	$-m_h/m$	γ_1^L	γ_2^L	κ^L	$-m_{so}/m$	g_c+g_{so}
Walton *et al.* (Ref. 29)	0.068 ± 0.015	0.50 ± 0.02	8.4	3.2	1.9	0.133	−7.6
Vrehen (Ref. 17)	0.082 ± 0.006	0.45 ± 0.05	7.2	2.5	1.1	0.159	−4.4
Stradling (Ref. 30)	0.087 ± 0.005	0.475 ± 0.015	6.8	2.4	1.0	0.170	−4.0
Narita *et al.* (Ref. 28)	0.1	0.38	6.4	1.8	0.8	0.185	−3.3

Here γ_1^L and κ^L are parameters originally introduced by Luttinger,[27] and E_p is defined by

$$m/m_c = 1 + \tfrac{1}{3} E_p[2/\mathcal{E}_g + 1/(\mathcal{E}_g+\Delta)] \quad . \tag{9}$$

Equation (8) is the analog of Roth's relation[19] for the conduction-band g factor,

$$g_c = 2\{1 - (E_p/3\,\mathcal{E}_g)[\Delta/(\mathcal{E}_g+\Delta)]\} \quad . \tag{10}$$

This relation together with Eq. (8), yields a rather simple result for the sum of the two g factors

$$g_c + g_{so} = -4\kappa^L \quad . \tag{11}$$

This shows that our experiment provides a direct measurement of the parameter κ^L. In many cases κ^L is not treated as an independent parameter, but rather is obtained to a good approximation from a knowledge of γ_1^L and γ_2^L,

$$\kappa^L = \tfrac{1}{3}(5\gamma_2^L - \gamma_1^L - 2). \tag{12}$$

Here and in the following discussion we neglect anisotropy of the valence bands with respect to crystal orientation. This is justification in view of the failure of Vrehen's magnetoabsorption experiments[17] to detect any such anisotropy. In terms of Luttinger's theory,[27] this means $\gamma_2^L \approx \gamma_3^L$. Finally, the light- and heavy-hole band effective masses are related to γ_1^L and γ_2^L:

$$\begin{aligned} -m/m_l &= \gamma_1^L + 2\gamma_2^L , \\ -m/m_h &= \gamma_1^L - 2\gamma_2^L . \end{aligned} \tag{13}$$

Values for γ_1^L, γ_2^L, and κ^L have been obtained from interband magnetoabsorption by Vrehen[17] and Narita.[28] They are listed in Table I along with the effective masses calculated using Eqs. (13). Narita treated κ^L as an independent parameter, whereas Vrehen used the approximate relation in Eq. (12). Values for the effective masses m_l and m_h have been determined by Walton and Mishra[29] from reflectivity and Faraday-effect measurements and by Stradling[30] from cyclotron resonance experiments. These values, together with the values for γ_1^L, γ_2^L, and κ^L calculated from Eqs. (12) and (13), are also given in Table I. In the last two columns we show the values for m_{so} and (g_c+g_{so}) calculated using Eqs. (7) and (11) for each set of experimental values. We used $m_c = 0.0665m$, $\Delta = 0.341$ eV, and $\mathcal{E}_g = 1.519$ eV. From Eq. (9), we calculated $E_p = 22.3$ eV, and the second term on the right-hand side of Eq. (7) turned out to be 0.90. From Table I we see that our experimental results, $m_{so} = -(0.154 \pm 0.005)m$ and $(g_c+g_{so}) = -4.7 \pm 1.0$, are in very good agreement with the values predicted from Vrehen's results.

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