Γ_1 conduction electron g factor and matrix elements in GaAs and $Al_xGa_{1-x}As$ alloys

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(Received 23 June 1975)

We report a new technique for determining $|g^*|$ of Γ_1 conduction electrons using optical-spin orientation techniques (polarization of luminescence, Hanle effect, and lifetime measurements). The variation of $|g^*|$ with x in $Al_xGa_{1-x}As$ ($0 \leq x \leq 0.21$) implies a negative g^* in GaAs, in contrast to previous theoretical interpretations but in agreement with various recent measurements. The experimental values of g^* and m^* in GaAs are used to determine the momentum matrix elements connecting the Γ_1 state to the Γ_{15} valence and conduction states. The results obtained for GaAs are employed to calculate matrix elements and g factors in $Al_xGa_{1-x}As$ alloys and in a number of zinc-blende crystals. Improved agreement between the calculated and measured values of g^* is obtained in nearly all cases.

I. INTRODUCTION

The experimental measurement of the Γ_1 conduction-electron (Γ_{1c}) g factor in semiconductors provides a sensitive test of the theoretically determined momentum matrix elements between the various states at Γ. Previously calculated matrix elements, however, give g factors which are generally not in very good agreement with experiment.[1-4] In particular, theoretical calculations[5,6] and some measurements[7,8] have led to the assignment of a positive value to g^* for GaAs. Two different experimental measurements, however, one by Weisbuch[1] and the other presented here, on the variation of g^* with alloying indicate a negative g factor for GaAs. Our results are in satisfactory agreement with the negative sign and magnitude of g^* obtained from measurements of the Zeeman spectrum of the photoluminescence emission line due to the recombination of excitons bound to neutral Sn acceptors[9] and from spin-flip Raman scattering.[10] Our experiment utilizes spin polarization and Hanle effect combined with measurement of minority-carrier lifetime to determine g^*. The details of the experiment are discussed in Sec. II.

We have used the measured value of g^* in GaAs in conjunction with effective-mass and optical data to determine the momentum matrix elements between the Γ_{1c} state and the Γ_{15} valence and conduction states. We find that the Γ_{15} conduction state makes an appreciable contribution to *both* g^* and m^* of the Γ_{1c} state in GaAs. The information on the momentum matrix element connecting the Γ_{15c} and Γ_{1c} states obtained from GaAs allows a calculation of g^* in $Al_xGa_{1-x}As$ alloys and some other zinc-blende crystals. The calculations and results are presented in Sec. III.

II. DISCUSSION OF EXPERIMENT

The determination of conduction-electron g factors in semiconductors by conventional EPR techniques is limited to systems in which sufficient equilibrium concentration of electrons is available (e.g., by adding donors), and then interpretation is sometimes complicated because of the presence of these donors. Recently, optical spin-orientation techniques have been used to measure $|g^*|$ in GaSb,[2] GaAs,[3] and InP.[3,4] It is found that $|g^*| = 0.51$ for pure GaAs,[3] and $|g^*| = 0.56$ for $In_{0.02}Ga_{0.98}As$.[1] (See note added in proof.) This indicates that the sign of g^* is negative for GaAs, because the addition of In lowers the energy gap and the effective mass, both of which tend to make g^* more negative.[11] We have used a different experimental technique (described below) to measure g^* in $Al_xGa_{1-x}As$ alloys and our results confirm the assignment of a negative value to g^* in GaAs.

To determine $|g^*|$ for $Al_xGa_{1-x}As$ ($0 \leq x \leq 0.21$) we have used spin polarization and Hanle effect combined with measurement of minority-carrier lifetime. Although it is more straightforward and precise to measure $|g^*|$ directly from the resonant decrease in the polarization, the small value of T_1 (about 1 nsec or less) in this system at low temperatures has thus far prevented us from seeing the resonance. This enhanced spin relaxation at low temperatures is apparently due to nuclear hyperfine effects.[13] The technique reported here, although less precise, is probably applicable to a wider range of materials.

The samples were produced by liquid-phase epitaxy on GaAs and contained about 10^{17}-cm^{-3} zinc acceptors [one of the pure GaAs samples was a bulk single crystal containing (2×10^{18})-cm^{-3} zinc acceptors]. Most samples were covered with a second epitaxial layer containing a high aluminum concentration ($x \approx 0.60$). Samples with such a window had minority-carrier lifetimes $\tau \approx 3$–10 nsec at 77 K. Samples for which the window was removed by mechanical polishing had $\tau \approx 0.2$–1 nsec.

The spin polarization was determined by optically pumping the samples with circularly polarized light from a cw krypton laser, using a pump photon energy about 50–100 meV larger than E_0, and mea-

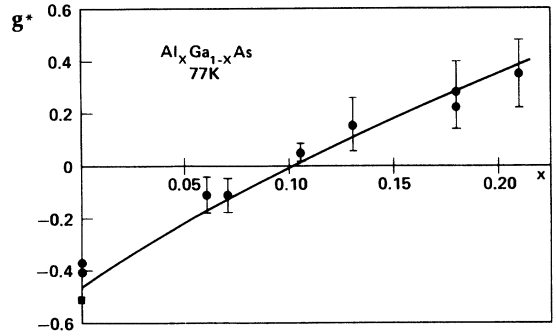

FIG. 1. g^* vs x for $Al_xGa_{1-x}As$. The measurement indicated by ■ is from Weisbuch (Ref. 1). The solid line is the theoretical result.

suring the degree of circular polarization of the luminescence. It is well known[14] that for a cubic semiconductor this polarization is related to τ and T_1 by

$$\rho = \tfrac{1}{4}\, T_1/(\tau + T_1) \ . \tag{1}$$

In the presence of a transverse magnetic field, ρ decreases (the Hanle effect) according to[14]

$$\rho(B) = \rho(0)/(1 + \omega_L T') \ , \tag{2}$$

where $\omega_L = g^* eB/2m$ and $T' = (\tau^{-1} + T_1^{-1})^{-1}$.

The lifetime τ was measured by mode locking the laser and using the phase-shift method.[15] Using a vector voltmeter, we measure the phase shift to within about 3°, which translates to a useful range of about 0.2–10 nsec for measuring τ. We estimate that the uncertainty in τ is about a factor of 2 at these two extremes and is about 10% for $\tau = 1$ nsec. The decay process is assumed to be exponential.

Thus the measurement of τ and T_1 enables us to determine g^* from the Hanle effect. Our results for $Al_xGa_{1-x}As$ are shown in Fig. 1. The uncertainties shown are primarily due to uncertainty in τ. Of course this measurement does not determine the sign of g^*. The sign was chosen to give a monotonically increasing variation with x, which in turn indicates a negative g^* for GaAs.

III. MATRIX ELEMENTS AT Γ

The effective gyromagnetic ratio g^* of the Γ_{1c} electrons can be evaluated from[11]

$$g^* = 2 - \frac{2i}{m} \sum_{n \neq \alpha} \frac{p_{\alpha n}^x p_{n\alpha}^y - p_{\alpha n}^y p_{n\alpha}^x}{E - E_n} \ , \tag{3}$$

where $|\alpha\rangle = |\Gamma_{1c}\rangle$ and $p_{\alpha n}$ represents a momentum matrix element. In zinc-blende crystals $p_{\alpha n}$ is nonzero only when n is a wave function with Γ_{15} symmetry, and the sum in Eq. (3) is nonzero only in the presence of spin-orbit interactions. The most important contributions to g^* (and similarly for the effective mass m^*) arise from the Γ_{15v} and Γ_{15c} states (shown in Fig. 2), and Eq. (3) leads to[11]

$$g^* = 2 - \tfrac{2}{3} P^2 \left(\frac{\Delta_0}{E_0(E_0 + \Delta_0)} + \lambda^2 \frac{\Delta_0'}{(E_0' - E_0)(E_0' - E_0 - \Delta_0')} \right) , \tag{4}$$

where

$$P^2 = 2/m \left| \langle \Gamma_{15v,x} | p_x | \Gamma_{1c} \rangle \right|^2$$

and

$$\lambda^2 P^2 = 2/m \left| \langle \Gamma_{15c,x} | p_x | \Gamma_{1c} \rangle \right|^2 \ .$$

In group-IV crystals where both the lowest conduction state ($\Gamma_{2'}$) and the Γ_{15c} state have odd parities, the matrix element $\langle \Gamma_{15c} | \vec{p} | \Gamma_{2'c} \rangle$ is zero (i.e., $\lambda = 0$) and only the states at the top of the valence band contribute to g^*. In zinc-blende crystals λ is different from zero as a result of the mixing, caused by the antisymmetric potentials,[12] of even and odd parity states at Γ.

The matrix element P^2 or $\lambda^2 P^2$ in Eq. (4) can be eliminated by using the effective-mass equation

$$\frac{m}{m^*} \simeq 1 + \frac{P^2}{3} \left(\frac{3E_0 + 2\Delta_0}{E_0(E_0 + \Delta_0)} - \lambda^2 \frac{3(E_0' - E_0) - 2\Delta_0'}{(E_0' - E_0)(E_0' - E_0 - \Delta_0')} \right) . \tag{5}$$

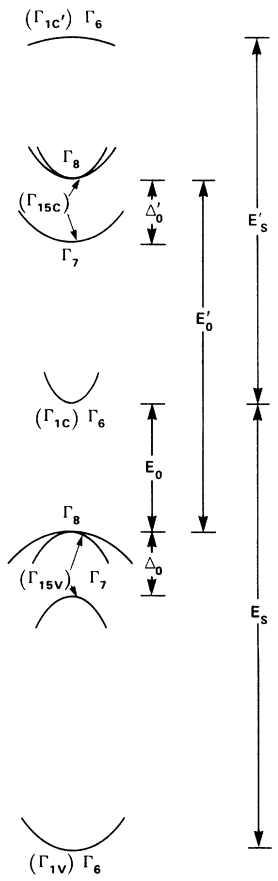

FIG. 2. Energy levels at Γ for zinc-blende crystals.

TABLE I. Calculated and experimental values of g^* are compared for a number of zinc-blende crystals. The calculated values of g^* are obtained from Eqs. (4) and (5) using $\lambda^2 = 0.4$ together with effective-mass and band-parameter data listed above.

	m^*/m (Ref. 24)	E_0 (Ref. 24)	Δ_0	E_0'	Δ_0'	P^2	g^* (calc.)	g^* (expt.)
GaAs	0.066	1.52	0.341[a]	4.659[a]	0.171[a]	29	−0.47	−0.51[b] ± 0.03
GaSb	0.045	0.81	0.80[c]	3.69[d]	0.25[e]	23.93	−8.0	−9.3[f] ± 0.3
InP	0.080[g]	1.42	0.11[h]	5.66[e]	0.41[e]	19.50	+1.21	1.26 ± 0.05[d]
InAs	0.024	0.42	0.38[j]	4.58[d]	0.43	21.36	−14.26	−14.7[k]
InSb	0.0137	0.237	0.81[j]	3.78[d]	0.41[e]	23.89	−50.23	−51.4[l] ± 0.1

[a]See Ref. 16.
[b]See Refs. 3, 4, 9, and 10.
[c]See Ref. 22.
[d]See Ref. 25.
[e]See Ref. 26.
[f]See Ref. 2.
[g]See Ref. 17.
[h]See Ref. 27.
[i]See Refs. 3 and 4.
[j]See Ref. 24.
[k]See Ref. 28.
[l]See Ref. 29.

For $\lambda = 0$ the resulting expression is identical to that obtained originally by Roth *et al.*[11] For GaAs assuming $\lambda = 0$ and using (see Fig. 2) $E_0 = 1.518$ eV, $\Delta_0 = 0.341$ eV, $E_0' = 4.659$ eV, $\Delta_0' = 0.171$ eV,[16] and $m^*/m = 0.066$,[17] for the Γ_{1c} state, Eqs. (4) and (5) yield $g^* = 0.16$, in poor agreement with the experimental value of -0.51. This clearly indicates the need for including the contribution of the Γ_{15c} states to m^* and g^*. Cardona[5] has used an expression equivalent to Eq. (4) to calculate g^* in a number of semiconductors. In evaluating λ he takes into account the mixing of even- and odd-parity states for the Γ_{15v} and Γ_{15c} states, but not for the Γ_{1c} state. This allows a simple evaluation of λ^2 from the measured $E_0' = E(\Gamma_{15c}) - E(\Gamma_{15v})$ gaps in GaAs and Ge. In this way Cardona obtains $\lambda^2 \simeq 0.19$ for GaAs which, using Eqs. (4) and (5), yields[18] $g^* = +0.09$. This is an improvement over the result for $\lambda^2 = 0$ but still not in agreement with the experimental value of -0.51. It should be noted that a higher value for λ^2 decreases g^* in two different ways: (i) through the term involving λ^2 in Eq. (4) and (ii) mainly through the increase in the value of P^2 resulting from Eq. (5), i.e., when λ^2 becomes larger P^2 also increases so that m^* is unchanged. Values of $\lambda^2 \simeq 0.4$ and $P^2 \simeq 29$ eV are necessary to explain *both* m^* and g^* in GaAs.

We have used the empirical pseudopotential method[19] to calculate the energy levels, wave functions, and matrix elements at Γ for GaAs. We find, however, that the calculations with both local and nonlocal (angular-dependent) potentials give values for λ^2 and P^2 that are much smaller than needed to explain m^* and g^*. Since $\lambda^2 = 0$ in diamond-structure crystals and $\lambda^2 \neq 0$ in zinc-blende crystals one may expect, on the basis of arguments similar to that used by Cardona,[5] that λ^2 depends primarily on the ionicity of the bond and that it should, therefore, be nearly equal in zinc-blende crystals with nearly the same ionicities. Therefore, it is possible to test whether $\lambda^2 = 0.4$ is reasonable in GaAs by using the same value of λ^2 for InAs, GaSb, InSb, and InP to calculate P^2 [from Eq. (5) using effective-mass and optical data] and g^*. These zinc-blende crystals have approximately the same ionicities on the Phillips–Van Vechten scale.[20] The results of such a calculation are shown in Table I, and the agreement between the calculated and experimental values is, in general, extremely good. Another test is provided by the $Al_xGa_{1-x}As$ alloys. For this system the increase of E_0 and decrease of Δ_0 with x will cause g^* to increase. The band parameters for $Al_xGa_{1-x}As$ were obtained from the data of Berolo and Woolley.[21,22] For $0 \leq x \leq 0.21$ E_0 varies between 1.518 and 0.316 eV. The parameter P^2 varies from 29 eV in GaAs to 21.1 eV in AlAs.[23] The variation of the term involving λ^2 in Eq. (4) with x was ignored since it produced only a very small variation in g^*. The parameter λ^2 was taken equal to 0.4 and a linear variation of band parameters with x was assumed. The result is shown as the solid line in Fig. 1 and it is seen that the agreement with experiment is very good.

The good agreement between the calculated and experimental g factors for $Al_xGa_{1-x}As$ alloys and also for the zinc-blende crystals listed in Table I shows that the assumption $\lambda^2 \simeq 0.4$ is reasonable in these systems. It is also interesting to look at the parameter

$$F' = -\lambda^2 P^2/(E_0' - E_0) , \tag{6}$$

which gives the contribution of the Γ_{15c} states to the effective mass m^* of the Γ_{1c} state. Lawaetz,[23] from an over-all fit to the experimental data, assumes that the total contribution of all Γ_{15c} symmetry bands to m^* can be represented by $F' \simeq -2$. Substituting the parameters listed in Table I and $\lambda^2 = 0.4$ in Eq. (6), we find F' equal to -3.70 in GaAs, -3.32 in GaSb, -1.84 in InP, -2.05 in InAs, and -2.70 in InSb. The use of a constant

versus a varying F' gives rise to differences of about 10% in values for P^2 obtained from Eq. (5).

In conclusion, we have shown that the variation of g^* in $Al_xGa_{1-x}As$ is consistent with the assignment of a negative g^* to GaAs. Furthermore, we have used the g-factor and effective-mass data to determine some momentum matrix elements at Γ in a number of zinc-blende crystals. For GaAs and GaSb we find the coupling of the Γ_{1c} state to higher conduction states (in particular the Γ_{15c} state) to be (70–80)% stronger than previously estimated.[23] Our results for the matrix elements P^2 between the Γ_{1c} and Γ_{15v} states agree to within 10% with those obtained by Lawaetz.[23]

ACKNOWLEDGMENTS

We are grateful to Dr. C. Weisbuch for communicating results prior to publication and for useful discussions. We also profited from discussions with J. B. Boyce, R. M. Martin, and R. M. White. A. Alimonda assisted with sample preparation.

Note added in proof: Dr. C. Weisbuch has informed us of the results of new measurements of the g factor of GaAs. The new experiments were performed to study the influence of the nuclear polarization on the measured g factor. The nuclear effects were found to be non-negligible. To cancel the nuclear fields, electron spin resonance and nuclear resonance (to depolarize the nuclei) were carried out simultaneously. The new values obtained for the g factors are: $|g^*| = 0.44$ in GaAs and $|g^*| = 0.46$ in $Ga_{0.99}In_{0.01}As$. As discussed in Sec. II these results indicate a negative g factor for GaAs. Using the new value of g^* in GaAs together with $m^*/m = 0.066$ for the Γ_{1c} electron we find that Eqs. (4) and (5) give $P^2 = 28.7$ eV and $\lambda^2 = 0.38$ in close agreement to the values of 29 and 0.4 which we have used in this paper.

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