

to be reduced by the Heisenberg spin-flip exchange energy (which approaches zero as  $U_{EE} \rightarrow \infty$ ),<sup>6</sup> since the electron exchanges which occur during hole migration may be regarded as multi-magnon excitations which will become inelastic for finite  $U_{EE}$ , thus reducing their contribution to the exciton line broadening. It seems likely, however, that the intraorbital  $U_{EE}$  will normally be larger than the intraorbital  $U_{TE}$  so that the limit discussed ( $U_{EE} \gg U_{TE}$ ) will provide a reasonable qualitative guide. In real materials the hole can also tunnel directly to a second-neighbor metal-ion site via hybridization with the nonmetal orbitals, thus avoiding the spin restriction studied above.<sup>7</sup> This might also lead to AF exciton line shapes closer to a  $\delta$  function. Direct measurements, perhaps comparing Raman scattering from exciton states above and below  $T_N$  in order to eliminate backgrounds, should help clarify which of the mechanisms dominates hole propagation in real materials. In their study of the direct optical spectrum of NiO, Newman and Chrenko<sup>5</sup> did not observe dramatic changes in exciton line shape on heating through  $T_N$ .<sup>8</sup> The above calculations suggest (assuming that the simple cubic lattice gives a qualitative indication for the fcc case) that more dramatic changes would be expected in AF crystals such as NiF<sub>2</sub>.

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†On leave from Groupe de Physique des Solides, Ecole Normale Supérieure, Université de Paris, Paris VII, France.

‡On leave from Baker Laboratory, Cornell University, Ithaca, N. Y. 14850, as a John Guggenheim Memorial Fellow.

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## Parallel Nature of the $\Lambda_1$ - $\Lambda_3$ Energy Bands in Germanium\*

Stephen Koeppen, Paul Handler, and Stephen Jasperson†

*Physics Department and Materials Research Laboratory, University of Illinois, Urbana, Illinois 61801*

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We report that the 2-eV electroreflectance structures in germanium can be represented by a two-dimensional critical line which contains the  $L$  point and extends over approximately 80% of the distance from  $L$  to  $\Gamma$ .

Since 1962, when calculations of Brust, Phillips, and Bassani<sup>1</sup> indicated the probable existence of  $M_1$  critical points along the (111) symmetry direction in the Brillouin zone (BZ) of Ge, several workers<sup>2</sup> have found tentative experimental evidence supporting the assignment of the 2.1–2.3-eV transitions in Ge (designated  $E_1$  and  $E_1 + \Delta_1$  in this and all other diamond–zinc-blende semiconductors) to three-dimensional  $M_1$  critical points in the zone interior. The apparent nonoccurrence<sup>2–4</sup> of transitions originating at the  $L$

point on the BZ boundary has remained a mystery. Because the  $\Lambda_1$  and  $\Lambda_3$  energy bands are so nearly parallel over a large part of the BZ,<sup>5</sup> it seems more reasonable to suggest that the observed spectra may best be described by the two-dimensional critical-point theory.<sup>6</sup>

The present experiments were performed at room temperature using the electrolyte technique,<sup>7,8</sup> with modulations from flat band to depletion or flat band to accumulation. It must be emphasized that knowledge of the flat-band position,

which we were able to monitor under actual experimental conditions, *is necessary* to obtain data *which can be interpreted*. Experimental data which do not satisfy this criterion cannot be used to validate theoretical models. In addition, two other interrelated conditions must be met in order to use the simple, one-electron theory: The electric field must be large and the sample doping must be chosen such that the uniform-field approximation is valid.<sup>9</sup>

**Flat-band to depletion modulation.**—Figure 1(a) shows  $\Delta R/R$  obtained from a 0.1- $\Omega$ -cm *n*-type sample at room temperature with a flat-band-depletion modulation of 1.5 V. The magnitude of the electric field,  $8.1 \times 10^4$  V/cm, was obtained from data taken on the same sample under identical modulation conditions at 1.09 eV.<sup>7</sup> Figures 1(b) and 1(c) show  $\Delta\epsilon_1$  and  $\Delta\epsilon_2$  obtained from a Kramers-Kronig analysis of the data in Fig. 1(a). The curve for  $\Delta\epsilon_2$  clearly shows that the structure is a spin-orbit-split pair, separated by

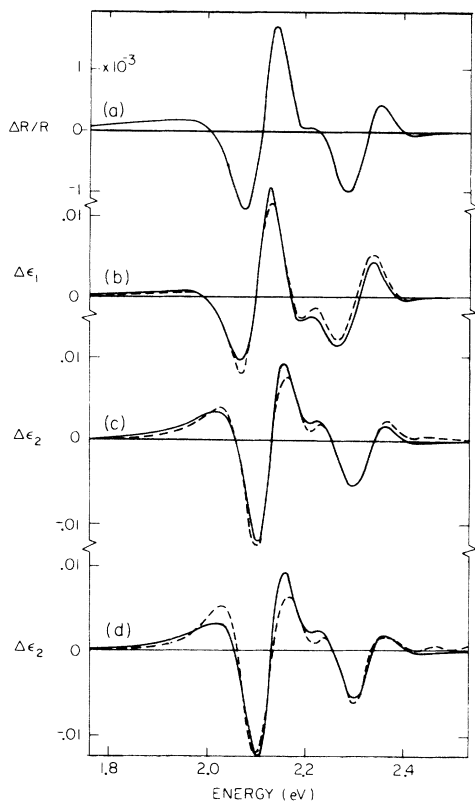


FIG. 1. The solid curves are (a)  $\Delta R/R$ , (b)  $\Delta\epsilon_1$ , and (c), (d)  $\Delta\epsilon_2$  obtained from a 0.1- $\Omega$ -cm *n*-type sample at a flat-band-depletion modulation of 1.5 V. The dashed curves in (b) and (c) are the results of a least-squares fit with the 2D  $M_0$  theory using Eq. (1). The dashed curve in (d) is the best fit by the 3D  $M_0$  theory.

about 0.2 eV. The positive values of the leading edges in  $\Delta\epsilon_1$  and  $\Delta\epsilon_2$  eliminate all possibilities except  $M_0$  critical points,<sup>10</sup> including the commonly accepted three-dimensional (3D)  $M_1$ . Since the 1D  $M_0$  is precluded by the symmetry along the  $\Lambda$  direction, this leaves only the 2D and 3D  $M_0$  theories with which to compare our data.

The dashed lines in Figs. 1(b) and 1(c) were calculated from the following equations:

$$\begin{pmatrix} \Delta\epsilon_1 \\ \Delta\epsilon_2 \end{pmatrix} = \frac{A}{(\hbar\omega)^2} \left[ \begin{pmatrix} G_2(\eta_1, \Gamma_1) \\ F_2(\eta_1, \Gamma_1) \end{pmatrix} + \begin{pmatrix} G_2(\eta_2, \Gamma_2) \\ F_2(\eta_2, \Gamma_2) \end{pmatrix} \right], \quad (1)$$

where  $F_2$  and  $G_2$  are the two-dimensional electro-optic functions,<sup>10</sup>  $A$  is an amplitude factor,  $\eta_i = (\hbar\omega - E_i)/\hbar\theta$ ,  $\Gamma_i = \gamma_i/\hbar\theta$ , and  $\theta = (e^2 \mathcal{E}^2 / 2\mu_F \hbar)^{1/3}$ .  $E_i$  and  $\gamma_i$  are the energy gap and broadening parameter, respectively, for the  $i$ th structure ( $i = 1, 2$ ). A single amplitude  $A$  and reduced mass in the direction of the field  $\mu_F$  were used for both structures since the large broadening, and the resultant uncertainty in  $A$  and  $\mu_F$ , made it meaningless to try to evaluate any (slight) difference in the  $A$ 's or  $\mu_F$ 's. In addition, although the broadening seems to be varying quite rapidly in this spectral region, two constant energy-independent  $\gamma$ 's, one for each structure, were used in the fit. The values obtained from the fit of Eq. (1) to the data were  $A = 26.5$ ,  $E_1 = 2.087$  eV,  $E_2 = 2.291$  eV,  $\gamma_1 = 0.053$  eV, and  $\gamma_2 = 0.069$  eV. *These same parameters* fit our data for several electric fields in the range  $(2-8) \times 10^4$  V/cm although the best fit occurs at the highest field as expected. From the known value of the electric field, and  $\hbar\theta = 0.044$  eV obtained from the fit, the transverse reduced mass was determined to be  $\mu_t = 0.020^{+0.024}_{-0.012}$ . If we use the known value of the transverse electron mass at the  $L$  point, 0.082,<sup>5</sup> this gives a value for the transverse hole mass of  $m_{ht}^* = 0.027^{+0.068}_{-0.018}$  as compared to the value of 0.12 given in Ref. 5 although no uncertainty was given in that reference. Our uncertainty was assumed to result from the uncertainty in the energy resolution of the data,  $\pm 5$  meV. Because of the nonlinear relation between peak widths and  $\mu_F$ , this translates into an asymmetric uncertainty in the masses.

The dashed curve in Fig. 1(d) was calculated from the 3D  $M_0$  theory using Eq. (1), but with the three-dimensional electro-optic functions. The parameters obtained from this fit were  $A = 12.8$ ,  $E_1 = 2.065$  eV,  $E_2 = 2.266$  eV,  $\gamma_1 = 0.038$  eV,  $\gamma_2 = 0.052$  eV, and  $\hbar\theta = 0.053$  eV. Since the longitudinal reduced mass is considerably larger than

the transverse reduced mass,  $\hbar\theta$  is determined by the latter, and we find a value  $\mu_t = 0.012^{+0.020}_{-0.005}$  which gives a transverse hole mass of  $m_{ht}^* = 0.014^{+0.038}_{-0.007}$ .

The agreement between experiment and the 2D  $M_0$  theory in Figs. 1(b) and 1(c) is excellent. Although the 3D  $M_0$  theory of Fig. 1(d) fits the data fairly well, the 2D fit is superior and in addition gives a transverse hole mass which is closer to that predicted by theory.

The most conclusive evidence of the two-dimensional nature of these transitions comes from the large amplitude factor. The expression for the 3D amplitude is<sup>11</sup>

$$A_{3D} = 4\sqrt{2} e^2 |\hat{\alpha} \cdot \vec{p}|^2 \mu_t \mu_i^{1/2} (\hbar\theta)^{1/2} / \hbar m^{1/2}, \quad (\text{cgs units}), \quad (2)$$

where  $\hat{\alpha}$  is the direction of polarization of the light. For a (100) face crystal, summing  $|\hat{\alpha} \cdot \vec{p}|^2$  over the eight equivalent  $\langle 111 \rangle$  directions gives a value  $\frac{16}{3} p_i^2$  ( $p_i \approx 0$ ) so that

$$A_{3D} = 64\sqrt{2} e^2 p_i^2 \mu_t \mu_i^{1/2} (\hbar\theta)^{1/2} / 3\hbar m^{1/2}, \quad (\text{cgs units}). \quad (3)$$

If we now take the value of  $A_{3D}$  and  $\mu_t$  obtained above from the fit to experiment together with the one-electron matrix element  $p_i = 0.65$  a.u.,<sup>12</sup> we find the longitudinal reduced mass to be  $\mu_i = 7.9$ . This is an extremely large reduced mass and is in fact too large for the 3D theory to be valid since it requires the bands to be parabolic over a region of  $k$  space along the  $\Lambda$  direction much greater than the width of the BZ. We therefore prefer the 2D theory since the valence and conduction bands are very nearly parallel along this direction. An estimate of the extent of the 2D region can be made in the following way. By limiting the density-of-states integration along the  $[111]$  direction to a finite region of  $k$  space,  $\Delta K_{111}$ , we find the expression for the 2D amplitude to be

$$A_{2D} = 32e^2 p_i^2 \mu_t \Delta K_{111} / 3m, \quad (\text{cgs units}), \quad (4)$$

which includes the sum over the eight equivalent  $\langle 111 \rangle$  directions. Using the value of  $A_{2D}$  and  $\mu_t$  obtained above and the one-electron value for  $p_i$ , we find  $\Delta K_{111} = 0.4$  a.u. This is about 80% of the distance between the  $L$  point and  $\Gamma$ , which is 0.51 a.u., and agrees very well with band-structure calculations.<sup>5</sup> Although the 2D theory is strictly correct only for bands which are perfectly parallel in one direction, it will be a good approxima-

tion when the broadening is comparable to or larger than the deviation from parallelism of the valence and conduction bands.

**Band filling.**—Since Ge is unique among semiconductors in that the minimum of the conduction band occurs at the  $L$  point, band filling<sup>13</sup> at 2 eV would imply that transitions near the  $L$  point are degenerate with transitions in the zone interior, formerly regarded to be solely responsible for the 2-eV structure, and would further support the 2D nature of this region.

We have observed band-filling effects by modulating the energy bands so that a degenerate accumulation layer is formed near the surface of the semiconductor. The occupation of states near the bottom of the conduction band should give rise to a decrease in  $\epsilon_2$  for transitions at and immediately above the gap energy. The solid curve in Fig. 2(a) shows  $\langle \Delta\epsilon_2 \rangle_{\text{eff}}$ <sup>14</sup> obtained from the data for a flat-band-accumulation modulation of 0.5 V. The points A, B, C, and D are the peak positions in energy of the first negative peak for flat-band-accumulation modulations of 0.1, 0.2, 0.3, and 0.5 V, respectively. The first positive and second negative peaks exhibit a similar shift

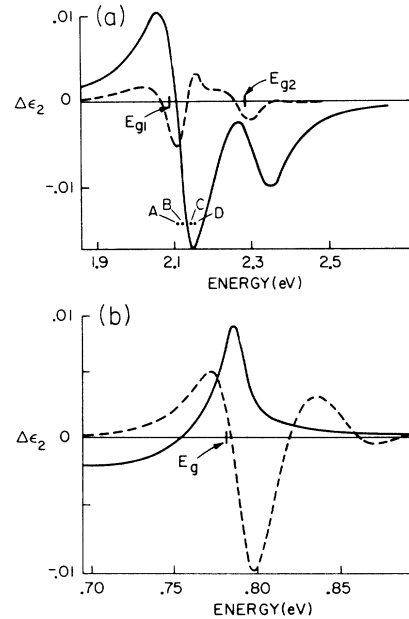


FIG. 2.  $\Delta\epsilon_2$  for a flat-band-accumulation modulation of 0.5 V (solid curves) and a flat-band-depletion modulation of 0.6 V (dashed curves) at (a) 2 eV and (b) the direct edge. The points A, B, C, and D in (a) are the positions in energy of the first negative peak for accumulation data with modulations from flat band of 0.1, 0.2, 0.3, and 0.5 V, respectively.

to higher energies as the modulation voltage increases.

In order to demonstrate that this observed accumulation line shape at 2 eV is indeed due to band filling and not due to the Franz-Keldysh effect, possibly modified by quantization or exciton screening, we compare data taken at both the direct edge (which is also an  $M_0$ -type transition) and 2 eV under the same modulation conditions. If no band filling were present at 2 eV, then the 2-eV and the direct-edge accumulation line shapes should be similar in nature since any quantization or screening should affect all transitions in a similar manner throughout the BZ.<sup>15</sup> Figure 2(b) shows direct-edge accumulation (solid curve) and depletion (dashed curve) line shapes taken under the same modulation conditions as the corresponding curves in Fig. 2(a). The point is that the depletion curves are similar in nature, characteristic of the Franz-Keldysh effect at  $M_0$ -type critical points, while the accumulation curves in Figs. 2(a) and 2(b) are *strikingly dissimilar*. The direct edge has a negative tail at lower energies while the 2-eV structure has a big positive peak. In addition, the direct-edge accumulation curve is the same order of magnitude as its depletion curve while the 2-eV accumulation curve is considerably larger than its depletion curve. We now show that the accumulation curves in Figs. 2(a) and 2(b) are in fact what one would expect from band filling and the Franz-Keldysh effect, respectively, in the presence of a large electric-field inhomogeneity.

In Fig. 3(a), we have plotted the uniform-field

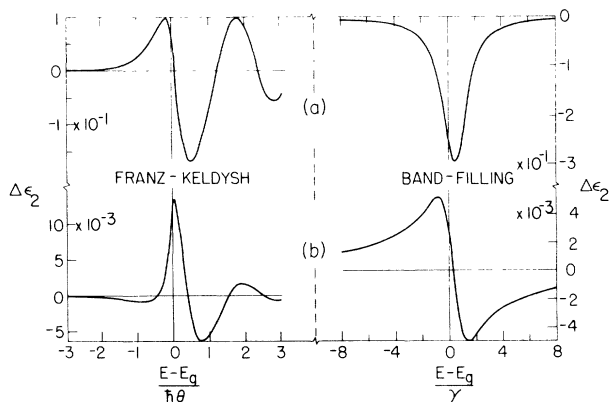


FIG. 3. Theoretical curves of  $\Delta\epsilon_2$  for the Franz-Keldysh and band-filling effects in (a) the uniform-field case and (b) the limit of large field nonuniformity. The Franz-Keldysh line shapes were calculated with  $\gamma/\hbar\theta = 0.1$  while the band-filling line shapes were calculated with  $\Delta/\gamma = 1.0$  (see text).

curves of  $\Delta\epsilon_2$  for the Franz-Keldysh effect, with a broadening of  $\gamma/\hbar\theta = 0.1$ ,<sup>7</sup> and for a simple two-dimensional band-filling model with  $\Delta/\gamma = 1.0$ , where  $\Delta$  is the distance the Fermi level is above bottom of the conduction band and  $\gamma$  is the broadening in energy units. Although this simple model for band filling is by no means exact, the broadening in this energy region is large enough so that it will at least give a qualitative picture of what the line shape should look like. Figure 3(b) shows  $\langle\Delta\epsilon_2\rangle_{eff}$  in the limit of a large field nonuniformity. The curves were calculated using the theory of Aspnes and Froya<sup>14</sup> and a value of  $R_1/2K = 10$ .<sup>9</sup> In accord with the results of Aspnes and Froya,<sup>14</sup>  $\langle\Delta\epsilon_2\rangle$  is similar to the negative of the uniform-field line shape for  $\Delta\epsilon_1$  while  $\langle\Delta\epsilon_1\rangle$  is similar to the uniform-field line shape for  $\Delta\epsilon_2$ . This is a useful rule to remember when working in the limit of extreme field inhomogeneity.

The similarity between the curves of Fig. 3(b) and the corresponding accumulation curves of Fig. 2 is striking, considering the approximations made. The inclusion of small quantization effects would result in an asymmetry in the peak sizes of the band-filling line shape of Fig. 3(b), resulting in an even better match to the accumulation curve of Fig. 2(a). The lack of subsidiary oscillations in the direct edge accumulation line shape is not presently understood.

In summary, the results of both depletion and accumulation E electroreflectance data demonstrate that the 2.1–2.3-eV structure in Ge can best be described as a two-dimensional  $M_0$  critical point which encompasses approximately 80% of the Brillouin zone along the  $\Lambda$  direction and includes the  $L$  point. The excellent agreement with the one-electron theory indicates that exciton effects are of little importance in the presence of high fields and large lifetime broadening. It appears that the past failure to quantitatively describe this structure was not the result of inadequacies in the one-electron theory, but rather the result of the use of data which did not satisfy the three conditions mentioned in the introduction.

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†Present address: Worcester Polytechnic Institute, Department of Physics, Worcester, Mass. 01609.

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seem to be present in the definitions on p. 180. The correct expressions are  $F_2(\eta) = Ai_1(\kappa\eta) - H(-\eta)$  and  $G_2(\eta) = Gi_1(\kappa\eta) + \pi^{-1} \ln|\eta| - C_0$ , where  $\kappa = 2^{2/3}$ ,  $Gi_1(x) = -\int_0^x Gi(t) dt$ , and  $C_0 = [\int Gi(t) dt]_0 - \pi^{-1} \ln \kappa$ .

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<sup>15</sup>In GaAs, where the minimum of the conduction band occurs at  $\Gamma$ , one should expect the opposite situation as in Ge. That is, one would expect to see band filling at  $\Gamma$  but not at the  $L$  point. Recent experiments on GaAs by Pond of this laboratory show line shapes similar to those of Fig. 2(a) at  $\Gamma$  and the usual Franz-Keldysh effect at  $L$ , supporting our hypothesis.

## Spectrum of the Reaction $^{208}\text{Pb}(n, \gamma)^{209}\text{Pb}$ and Semidirect Capture Theory\*

I. Bergqvist,† D. Drake, and D. K. McDaniels‡

Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico 87544

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Measurements of the spectrum of  $\gamma$  rays from the reaction  $^{208}\text{Pb}(n, \gamma)^{209}\text{Pb}$  have been made at incident neutron energies of 9.2, 11.2, and 13.2 MeV. The observed spectral shapes disagree strongly with the predictions of compound-nuclear theory and agree quite well with those predicted by semidirect (collective) capture theory.

The earliest theory of radiative nucleon capture involved the *compound-nuclear* reaction mechanism. This theory explained the capture of low-energy nucleons quite well. However, the situation for the capture of nucleons with energies greater than 4 or 5 MeV is not as satisfactory. Upon comparing the results of the theoretical calculations with experiment, particularly measurements of 14-MeV neutron capture cross sections, serious discrepancies were found.<sup>1</sup> The calculated magnitude of the total radiative cross sections was usually 1 or 2 orders of magnitude low for low-mass nuclei and falls below the observed values by 4 to 5 orders of magnitude for heavy nuclei. In order to explain these differences, several authors<sup>2-5</sup> proposed a reaction mechanism based on a *direct* capture process, in which an incident nucleon moving in the optical potential of the target nucleus plus nucleon makes a radiative transition to a lower state. Unfortunately, this mechanism leads to a cross section

about an order of magnitude lower than the lowest neutron capture cross section.<sup>6</sup> To overcome this, a collective *semidirect* capture mechanism was proposed.<sup>7-9</sup> This theory assumes that the incident nucleon is captured into a lower orbit with the excitation of the target nucleus into its giant-dipole resonance as an intermediate state from which it decays radiatively with a collective enhancement. Evidence is now beginning to accumulate favoring this process.<sup>10</sup>

It has recently been claimed that compound-nuclear theory can predict the right order of magnitude of the neutron capture cross section for intermediate energies around 10 MeV.<sup>11</sup> A sensitive test of the relative importance of the compound-nuclear process to this reaction is to measure the shape of the spectrum of  $\gamma$  rays emitted in the capture process.

The purpose of the present experiment was to investigate the spectrum from the reaction  $^{208}\text{Pb}(n, \gamma)^{209}\text{Pb}$  at several energies and to compare the